

MOLTEN SALT REACTOR SYSTEMS
Compilation of Literature (2000-2010)

K.L. Ramakumar
Former Director, Radiochemistry & Isotope Group
Bhabha Atomic Research Centre
Mumbai 400 085

Sequence of Chapters

Preface
Literature Referred
Executive Summary
Introduction
Projection of Energy Needs
Technology Challenges
Chemistry Aspects of MSRs
Physics Of Molten Salt Reactors
Safety Considerations
AHTR
Useful information

Preface

Current investigations being carried out and the concepts of molten salt reactors are entirely based on the molten salt experiment (MSRE) of ORNL, USA in 1960s and the design of a molten salt breeder reactor (MSBR) of early 1970s, again by ORNL. These investigations cover almost all aspects of reactor design including, fuel identification, composition, materials of construction, compatibility issues, reactor physics aspects, expected thermal power output etc. Even though the MSBR programme was terminated during 1970, research continued up to as long as 1976. India did contribute to this molten salt reactor concept during early period of 1970s. The research activities were basically focused on solubility studies of actinide fluorides in different salt mixtures.

Extensive investigations carried out and literature made available by ORNL in the open domain on MSRE and MSBR, triggered a re-look into the whole reactor concept. Molten Salt Reactor is a very attractive concept especially for the Thorium fuel cycle which allows nuclear energy production with a very low production of radiotoxic minor actinides. Its main characteristic is the strong coupling between neutronics and salt processing. Such nuclear reactors use a liquid fuel which is also the coolant. This compendium attempts to bring out salient features of MSRE and MSBR, the relevance of molten salt reactors for future energy security and different design concepts being discussed in the open literature by various countries. It should be made clear that no molten salt reactor is currently operating and all the reference given here describe the concepts and theoretical calculations carried out in support of feasibility of such a concept.

Literature referred

C. Renault, M. Hron, R. Konings and D.E. Holcomb, *the molten salt reactor (MSR) in generation iv: overview and perspectives*, GIF Symposium – Paris (France) – 9-10 September, 2009

S. Delpech, E. Merle-Lucotte, T. Auger, X. Doligez, D. Heuer, G. Picard, *MSFR: material issues and the effect of chemistry control*, GIF Symposium – Paris (France) – 9-10 September, 2009

Charles W. Forsberg, Per F. Peterson, HaiHua Zhao, *An Advanced Molten Salt Reactor Using High-Temperature Reactor Technology*, 2004 International Congress on Advances in Nuclear Power Plants (ICAPP 04)

Charles W. Forsberg, *Reactors with Molten Salts: Options and Missions*, Frederic Joliot & Otto Han Summer School on Nuclear Reactors "Physics, Fuels, and Systems" Cadarache, France August 25–September 3, 2004

Charles Forsberg, *The Advanced High-Temperature Reactor: High-Temperature Fuel, Molten Salt Coolant, and Liquid-Metal-Reactor Plant*, 1st International Conference on Innovative Nuclear Energy Systems for Sustainable Development of the World (COE INES-1) Tokyo Institute of Technology; Tokyo, Japan October 31–November 4, 2004

Charles W. Forsberg, *Carbon-Carbon-Composite Salt-Cooled Electric Space Reactors*, Proceedings of the Space Nuclear Conference 2005 San Diego, California, June 5-9, 2005

Charles W. Forsberg, *Molten-Salt-Reactor Technology Gaps*, Proceedings of ICAPP '06 Reno, NV USA, June 4–8, 2006 Paper 6295

M. S. Coops, D. F. Bowersox, *Nonaqueous Processing Methods*, LA-10169-MS, (1984)

C. F. Baes, Jr., *The Chemistry and Thermodynamics of Molten Salt Reactor Fuels*, Conf-730701

David F. Williams, Frederick J. Peretz, *characterization of the molten salt reactor experiment fuel and flush salts*, CONF-9606116-7, American Nuclear Society (ANS) Meeting in Reno, Nevada, June 16-20, 1996.

L. C. Cadwallader, G. R. Longhurst, *FLIBE use in fusion reactors: an initial safety assessment*, INEEL/EXT-99-00331 March 1999

P.R. Kasten, E.S. Bennett, H.F. Bauman, W.L. Carter, W.B. McDonald, R.C. Robertson, J.H. Westsik, *Summary of Molten-Salt Breeder Reactor Design Studies*. ORNL-TM-1467(1966)

E. L. Compere, S. S. Kirsliis, E. G. Bohlmann, F. F. Blankenship, W. R. Grimes, *Fission Product Behavior in the Molten Salt Reactor Experiment*, ORNL-4865 (1975)

Charles W. Forsberg, Per F. Peterson, Larry Ott, *The Advanced High-Temperature Reactor (AHTR) for Producing Hydrogen to Manufacture Liquid Fuels*, 2004 Americas Nuclear Energy Symposium ANS, Florida

O. Benes, C. Cabet, S. Delpech, P. Hosnedl, V. Ignatiev, R. Konings, D. Lecarpentier, O. Matal, E. Merle-Lucotte, C. Renault, J. Uhlir, *Assessment of Liquid Salts for Innovative Applications (ALISIA)* 2009

L. G. Alexander, *molten-salt fast reactors*, ANL-6792

S. Delpech, E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le-Brun, X. Doligez, G. Picard, *Reactor physics and reprocessing scheme for innovative molten salt reactor system*, J. Fluorine Chem. (2008), doi:10.1016/j.jfluchem.2008.07.009

E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le Brun, *Introduction to the Physics of Molten Salt Reactors*

D. F. Williams and L. M. Toth, *Chemical Considerations for the Selection of the Coolant for the Advanced High-Temperature Reactor*, ORNL/GEN4/LTR-05-011 (2005)

David LeBlanc, *Molten Salt Reactors: A New Vision for a Generation IV Concept*

JAMES A. LANE (Editor), *Aqueous Homogeneous Reactors*, Compilation Report, ORNL (1958)

FISA 2009, Seventh European Commission conference on *Euratom research and training in reactor systems*, Conference Proceedings 22 - 24 June 2009, Prague, Czech Republic

Charles W. Forsberg, Per F. Peterson, David F. Williams, *Practical Aspects of Liquid-Salt-Cooled Fast-Neutron Reactors*, Proceedings of ICAPP'05 Seoul, KOREA, May 15-19, 2005 Paper: 5643

Charles W. Forsberg, P. F. Peterson, R. A. Kochendarfer, *Design Options for the Advanced High-Temperature Reactor*, International Congress on the Advances in Nuclear Power Plants (ICAPP'08) Anaheim, California June 8-12, 2008

David LeBlanc, *Molten salt reactors: A new beginning for an old idea*, Nuclear Engineering and Design

H. G. MacPherson, *The Molten Salt Reactor Adventure*, Nuclear Science And Engineering: **90**, 374 (1985)

R.J.S. van't Eind, *Simulation of a fast molten salt reactor*, PNR-131-2011-009 July 2011

R. W. Moira, *Cost of electricity from Molten Salt Reactors (MSR)*, Nuclear Technology **138**, 93 (2002)

Charles W. Forsberg, *Molten Salt Reactors (MSRs)*, The Americas Nuclear Energy Symposium (ANES 2002) American Nuclear Society Miami, Florida October 16-18, 2002

D. F. Williams, L. M. Toth, K. T. Clarno, *Assessment of Candidate Molten Salt Coolants for the Advanced High-Temperature Reactor, (AHTR)*, ORNL/TM-2006/12 (2006)

D. F. Williams, *Assessment of Candidate Molten Salt Coolants for the NGNP/NHI Heat-Transfer Loop*, ORNL/TM-2006/69

David Samuel, *Molten Salt Coolants for High Temperature Reactors a Literature Summary of Key R&D Activities and Challenges*, IAEA Internship Report INPRO COOL (2009)

Jorge Abejón Orzáez, *Neutronics Analysis Of A Modified Pebble Bed Advanced High Temperature Reactor Thesis*, The Ohio State University 2009

C. Petroski, *Design of a 2400MW Liquid-Salt Cooled Flexible Conversion Ratio Reactor*, Master of Science in Nuclear Engineering at the Massachusetts Institute of Technology September 2008

E. Merle-Lucotte, X. Doligez, D. Heuer, M. Allibert, V. Ghetta, *Simulation Tools And New Developments Of The Molten Salt Fast Reactor*

D. E. Holcomb, G. F. Flanagan, B. W. Patton, J. C. Gehin, R. L. Howard, T. J. Harrison, *Fast Spectrum Molten Salt Reactor Options*, ORNL/TM-2011/105 (2011)

A. Mourougov, P.M. Bokov, *Potentialities of the fast spectrum molten salt reactor concept: REBUS-3700*, Energy Conversion and Management **47**, 2761 (2006)

Ondřej Beneš, *Thermodynamics of Molten Salts for Nuclear Applications*, Thesis, Institute Of Chemical Technology, Prague Faculty of Chemical Technology Department of Inorganic Chemistry JRC-ITU-TN-2008(40)

Kazuo Furukawa, Kazuto Arakawa, L. Berrin Erbay, Yasuhiko Ito, Yoshio Kato, Hanna Kiyavitskaya, Alfred Lecocq, Koshi Mitachi, Ralph Moir, Hiroo Numata, J. Paul Pleasant, Yuzuru Sato, Yoichiro Shimazu, Vadim A. Simonenco, **Din Dayal Sood**, Carlos Urban, Ritsuo Yoshioka, *A road map for the realization of global-scale thorium breeding fuel cycle by single molten-fluoride flow*, Energy Conversion and Management **49**, 1832 (2008)

Jan Uhlíř, *Chemistry and technology of Molten Salt Reactors – history and perspectives*, Journal of Nuclear Materials, **360**, 6 (2007)

Victor Ignatiev and Aleksander Merzlyakov, *Transport Properties Of Molten-Salt Reactor Fuel Mixtures: The Case Of Na, Li, Be/F And Li, Be, Th/F Salts*

William A Casino, Kirk Sorensen, Christopher A Whitener, *A Small Mobile Molten Salt Reactor (SM-MSR) for Underdeveloped Countries and Remote Locations*, Graduate Design Team Entry American Nuclear Society Student Design Competition April 25, 2007

Towards an Alternative Nuclear Future, Capturing thorium-fuelled ADSR energy technology for Britain, Report of the thorium energy amplifier association, Britain (2009-2010)

L. Mathieu, D. Heuer, R. Brissot, C. Garzenne, C. Le Brun, D. Lecarpentier, E. Liatard, J.-M. Loiseaux, O. Me´plan, E. Merle-Lucotte, A. Nuttin, E. Walle, J. Wilson, *The thorium molten salt reactor: Moving on from the MSBR*, Progress in Nuclear Energy 48 (2006) 664-679

Shisheng Wang, Andrei Rineiski, and Werner Maschek, *Development and Verification of the SIMMER-III Code for Molten Salt Reactors*

Pavel Soucek, Frantisek Lisy, Radka Tulackova, Jan Uhlir and Rudolf Mraz, *Development of Electrochemical Separation Methods in Molten LiF-NaF-KF for the Molten Salt Reactor Fuel Cycle*, Journal of Nuclear Science and Technology, **42**, 1017(2005)

S. Wang, A. Rineiski, W. Maschek, *Thermal Hydraulics Studies for a Molten Salt Burner Reactor*

S. Wang, M. Flad, A. Rineiski, W. Maschek, *Extension of the SIMMER-III Code for Analysing Molten Salt Reactors*

J. R. Hightower, Jr., *Process Technology for the Molten-Salt Reactor ^{233}U -Th Cycle*, ANS 1975 Winter Meeting, San Francisco, California, November 16-21, 1975

O. S. Feynberg, V. V. Ignatiev, *Neutronic and Fuel Cycle Consideration: From Single Stream to Two- fluid Th-U Molten Salt System*, Proceedings of the First ACSEPT International Workshop Lisbon, Portugal, 31 March – 2 April 2010

Elsa Merle-Lucotte, Ludovic Mathieu, Daniel Heuer, Véronique Ghetta, *Influence of the Processing and Salt Composition on the Thorium Molten Salt Reactor*

L. Mathieu, D. Heuer, E. Merle-Lucotte, R. Brissot, and C. Le Brun, E. Liatard, J.-M. Loiseaux, O. Méplan, A. Nuttin and D. Lecarpentier, *Possible Configurations for the Thorium Molten Salt Reactor and Advantages of the Fast Nonmoderated Version*, Nuclear Science and Engineering, **161**, 78 (2009)

Reinhard Koch and Andrey Myasnikov, *Incineration Of Transuranic Waste in A Molten Salt Reactor Based on the Thorium-Uranium-233 Fuel Cycle*

Jitka Žáková, *Analysis of an Advanced Graphite Moderated and Molten Salt Cooled High Temperature Reactor*, Master of science thesis, Department of Reactor Physics, Royal Institute of Technology, Stockholm, Sweden 2006

Ralph W. Moir and Edward Teller, *Thorium-Fueled Underground Power Plant Based on Molten Salt Technology*, NUCLEAR TECHNOLOGY, **151**, 334(2005)

Charles W. Forsberg, *Thermal- and Fast-Spectrum Molten Salt Reactors for Actinide Burning and Fuel Production*, GLOBAL 07: Advanced Nuclear Fuel Cycles, September 9–13, 2007 Boise, Idaho

Victor Ignatiev¹, Raul Zakirov¹, Konstantin Grebenkine, *Molten Salts as Possible Fuel Fluids for TRU Fuelled Systems*: ISTC #1606 APPROACH

E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le Brun, R. Brissot, E. Liatard, L. Mathieu, *The Thorium Molten Salt Reactor: Launching the Thorium Cycle While Closing the Current Fuel Cycle*

Elsa MERLE-LUCOTTE (LPSC/ENSPG), France, Merle@Lpsc.In2p3.Fr –Daniel HEUER, *Molten Salt Reactor: Deterministic Safety Evaluation*, ENC 2005

Elsa Merle-Lucotte, Daniel Heuer, Michel Allibert, Xavier Doligez, Véronique Ghetta, Christian Le Brun, *Optimization and simplification of the concept of non-moderated Thorium Molten Salt Reactor*, International Conference on the Physics of Reactors “Nuclear Power: A Sustainable Resource” Casino-Kursaal Conference Center, Interlaken, Switzerland, September 14-19, 2008

L. Mathieu, D. Heuer, A. Billebaud, R. Brissot, C. Garzenne, C. Le Brun, D. Lecarpentier, E. Liatard, J.-M. Loiseaux, O. Meplan, E. Merle-Lucotte And A. Nuttin, *Proposal for a Simplified Thorium Molten Salt Reactor*, Proceedings of GLOBAL 2005 Tsukuba, Japan, Oct 9-13, 2005

E. Merle-Lucotte, D. Heuer, C. Le Brun, L. Mathieu, R. Brissot, E. Liatard, O. Meplan, A. Nuttin, *Fast Thorium Molten Salt Reactors started with Plutonium*, Proceedings of ICAPP '06 Reno, NV USA, June 4-8, 2006

E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le Brun, L. Mathieu, R. Brissot, E. Liatard, *Optimized Transition from the Reactors of Second and Third Generations to the Thorium Molten Salt Reactor*, Proceedings of ICAPP 2007 Nice, France, May 13-18, 2007

E. Merle-Lucotte, D. Heuer, M. Allibert, X. Doligez, V. Ghetta, *Optimizing the Burning Efficiency and the Deployment Capacities of the Molten Salt Fast Reactor*, Proceedings of Global 2009 Paris, France, September 6-11, 2009

E. Merle-Lucotte, L. Mathieu, D. Heuer, J-M. Loiseaux, A. Billebaud, R. Brissot, S. David, C. Garzenne, O. Laulan, C. Le Brun, D. Lecarpentier, E. Liatard, O. Méplan, F. Michel-Sendis, A. Nuttin and F. Perdu, *Molten Salt Reactors and Possible Scenarios for Future Nuclear Power Deployment*, PHYSOR 2004 - The Physic of Fuel Cycles and Advanced Nuclear Systems: Global Developments Chicago, Illinois, April 25-29, 2004

E. Merle-Lucotte, D. Heuer, C. Le Brun, M. Allibert, V. Ghetta, L. Mathieu, R. Brissot, R. Chambon, E. Liatard, *The Non-Moderated TMSR, an Efficient Actinide Burner and a Very Promising Thorium-Based Breeder*, GLOBAL07

E. Merle-Lucotte, D. Heuer, M. Allibert, X. Doligez, V. Ghetta, *Minimizing the Fissile Inventory of The Molten Salt Fast Reactor*, Advances in Nuclear Fuel Management IV (ANFM 2009) Hilton Head Island, South Carolina, USA, April 12-15, 2009

Sylvie Delpech, Elsa Merle-Lucotte, Thierry Auger, Xavier Doligez, Daniel Heuer, Gérard Picard, *MSFR: Material issues and the effect of chemistry control*, Proceedings of the GIF Symposium 2009 Paris, France, September 9-10, 2009

L. Mathieu, D. Heuer, A. Nuttin, F. Perdu, A. Billebaud, R. Brissot, C. Le Brun, E. Liatard, J.-M. Loiseaux, O. Méplan and E. Merle-Lucotte, S. David, C. Garzenne, D. Lecarpentier, *Thorium Molten Salt Reactor : from high breeding to simplified reprocessing*, GLOBAL 03

S. Delpech, S. Jaskierowicz, G. Picard, E. Merle-Lucotte, D. Heuer, X. Doligez, *Innovative nuclear system based on liquid fuel*, Proceedings of ICAPP '09 Tokyo, Japan, May 10-14, 2009

Elsa Merle-Lucotte, Ludovic Mathieu, Daniel Heuer, Annick Billebaud, Roger Brissot, Christian Le Brun, Eric Liatard, Jean-Marie Loiseaux, Olivier Meplan, Alexis Nuttin, Jonathan Wilson, *Influence of the Reprocessing on Molten Salt Reactor Behaviour*, MS07

Xavier Doligez, Daniel Heuer, Elsa Merle Lucotte, Véronique Ghetta, Michel Allibert, Sylvie Delpech, Gérard Picard, *Thorium Molten Salt Reactor reprocessing unit: Characterization and influence on the core behaviour*, MS08

E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le Brun, R. Brissot, E. Liatard, L. Mathieu, *The Thorium Molten Salt Reactor: Launching the Thorium Cycle While Closing the Current Fuel Cycle*, TMSR-ENC-07

The Use of Thorium in Nuclear Power Reactors, USAEC Report, WASH 1097 (1969)

EXECUTIVE SUMMARY

Nuclear reactor types can be classified by power output and the peak temperatures of their coolants. Light water reactors (LWRs) are low-temperature, high-pressure reactors. Traditional fast reactors with solid fuel and cooled with liquid sodium operate at medium temperatures and low pressures. Two options exist for high-temperature reactor coolants: (1) high-pressure gases and (2) low-pressure liquids with boiling points above the peak coolant temperatures. MSRs are a type of high-temperature reactor.

Molten Salt Reactors (MSRs) are liquid-fueled reactors that can be used for production of electricity, actinide burning, production of hydrogen, and production of fissile fuels. Electricity production and waste burn-down are envisioned as the primary missions for the MSR. Fissile, fertile, and fission isotopes are dissolved in a high-temperature molten fluoride salt with a very high boiling point (1,400°C) that is both the reactor fuel and the coolant. The near-atmospheric-pressure molten fuel salt flows through the reactor core. The traditional MSR designs have a graphite core that results in a thermal to epithermal neutron spectrum. Alternative designs are now being explored with no reactor internals and a fast neutron spectrum. In the core, fission occurs within the flowing fuel salt that is heated to $\sim 700^{\circ}\text{C}$, which then flows into a primary heat exchanger where the heat is transferred to a secondary molten salt coolant. The fuel salt then flows back to the reactor core. The clean salt in the secondary heat transport system transfers the heat from the primary heat exchanger to a high-temperature Brayton cycle that converts the heat to electricity. The Brayton cycle (with or without a steam bottoming cycle) may use either nitrogen or helium as a working gas.

Two experimental MSRs built at Oak Ridge National Laboratory (ORNL) established the basic technology for the MSR. The first reactor was the 2.5 MWt Aircraft Reactor Experiment that in 1954 demonstrated peak operating temperatures up to 860°C . This was part of an effort to build a nuclear-powered military aircraft with the jet engines receiving heat from the MSR via an intermediate heat transport loop. This was followed in the 1960s by the Molten Salt Reactor Experiment, an 8 MWt reactor, to demonstrate key features required for a Molten Salt Breeder Reactor (MSBR).

The renewed interest in MSRs is a consequence of changing goals and new technologies. Russian and Organization for Economic Cooperation and Development studies have identified the MSR as a potential component of a closed fuel cycle to efficiently burn actinides because it offers the potential to reduce the long-term radiotoxicity of the wastes produced from production of electricity in other types of reactors. The use of liquid fuels avoids some of the technical difficulties (such as fuel fabrication) for burning actinides, a particularly difficult fuel fabrication

challenge for the intensely radioactive higher actinides. The traditional MSR has a thermal to epithermal neutron spectrum. Recent work in France and elsewhere has developed the concept of a fast-spectrum MSR. Unlike solid-fuel fast-spectrum reactors, the fast-spectrum MSR has the potential safety advantages of strong negative Doppler and void coefficients—traditionally the major safety challenges for solid-fuel fast reactors. There is a secondary interest in the MSR's use for hydrogen production because of the high-temperature capability.

Much of the ongoing R&D on molten fluoride salts is directly applicable to the MSR. Three include:

- (i) **Liquid-Salt Heat-Transport Systems:** Liquid salts are being investigated for transport of heat from MSR to hydrogen production systems. Liquid salts are one of two candidates for this task. In Europe, the French Atomic Energy Commission (CEA) is investigating the use of liquid-salt intermediate loops to replace sodium intermediate loops in sodium fast reactors to reduce costs and eliminate the potential for chemical reactions between the reactor coolant and the power cycle. These high-temperature heat-transport loops are also candidates for use in solar power towers, in-situ recovery of shale oil, and tertiary oil recovery.
- (ii) **Advanced High-Temperature Reactor (AHTR):** The AHTR is a solid fuel reactor that uses a clean liquid salt coolant to transfer heat from the solid reactor core to an intermediate heat exchanger. The intermediate heat-transport loop transfers the heat to a Brayton power cycle or a hydrogen production facility.
- (iii) **Fusion Reactors:** Liquid salts are major candidates for cooling inertial and magnetic fusion energy systems.

Features of MSRs

The reactor characteristics minimize the potential for accident initiation. Unlike solid-fuel reactors, MSRs operate at steady-state conditions with no change in the nuclear reactivity of the fuel as a function of time. Fuel is added as needed; consequently, the reactor has low excess nuclear reactivity. No excess fuel is needed at reactor startup to compensate for fuel depletion, and no excess reactivity is required to override xenon poisoning. No significant buildup of xenon occurs over time because the xenon gas continuously exits via the off-gas system. There is a negative temperature coefficient because increased temperatures lower the fuel-salt density and push fuel out of the reactor core. The fast-spectrum MSRs, unlike traditional solid-fuel fast reactors, have strong negative temperature and void coefficients because of fuel expelled from the core at higher temperatures—a unique safety advantage of this type of fast reactor. In normal operations, the control rods are fully removed from the reactor.

Decay Heat Cooling and Accident Management: Molten salt reactors use passive emergency core cooling systems that are radically different from those used in solid-fuel reactors. If the molten reactor fuel salt overheats, its thermal expansion causes it to overflow by gravity into an overflow weir. The fuel is then dumped to multiple critically safe storage tanks with passive decay-heat cooling systems. Freeze valves that open upon overheating of the salt can also be used to initiate core dump of fuel. Drains under the primary system also dump fuel salt to the storage tanks if a primary system leak occurs. This design approach allows very large reactors to be built with passive safety systems.

Many of the driving forces for an accident are reduced compared with those that exist for other reactors. Fission products (with the exception of xenon and krypton) and nuclear materials are highly soluble in the salt and will remain in the salt under both operating and anticipated accident conditions. The fission products that are not soluble (e.g., xenon and krypton) are continuously removed from the molten fuel salt, solidified, packaged, and stored in passively cooled storage vaults. There are no major stored energy sources within containment such as high-pressure fluids (helium and water) or reactive fluids (sodium). This reduces requirements for the containment.

Physics aspects

As noted, MSRs are fluid-fuel reactors. Such reactors have several reactor physics characteristics that are different from those of solid-fuel reactors.

Nuclear Reactivity: Negligible xenon effect occurs because xenon continuously escapes from the fuel salt into the off-gas system. There is no change in core reactivity with time because fuel is continuously added as required. The fuel inventory in the reactor core is coupled to the reactor temperature. An increase in reactor temperature reduces the fuel inventory by expansion of the fuel salt with less mass of fuel salt in the reactor core. For fast-spectrum MSRs, this results in the unusual characteristic for a large fast reactor of having a strong negative temperature and void coefficient: a major potential safety advantage.

Fissile Inventory: As a class, MSRs have very low fissile inventories compared with other reactors with similar neutron spectrums: (1) a low fuel-cycle fissile inventory exists outside the reactor system (no conventional spent nuclear fuel [SNF]), (2) little excess reactivity is required to compensate for burnup because fuel is added on-line, (3) direct heat deposition in the fuel/coolant allows high power densities, and (4) the high-absorption fission products, such as xenon, are continuously removed. As a consequence, thermal spectrum MSRs require <2 kg of fissile material per MWe to reach criticality compared with 3 to 5 kg/MWe for LWRs and over 25 kg/MWe for fast-spectrum reactors. This implies that the MSR has the potential to provide long-term, sustainable energy production while limiting the

global inventory of plutonium and minor actinides to a total quantity over an order of magnitude lower than solid-fuel reactors.

Burnup and Plutonium Isotopics: In an MSR, fuel is added incrementally to the liquid as required. No excess fuel and associated burnable absorbers are required. Selected fission products are removed from the molten salt and solidified as a waste form. Consequently, the normal burnup limits that define solid fuels do not apply to a liquid-fuel reactor. The plutonium remains in the salt, with the lighter plutonium fissile isotopes burned out faster than ^{242}Pu . This has major implications in terms of proliferation resistance. The high ^{242}Pu content makes the plutonium from an MSR much less desirable than plutonium from any other reactor type for use in weapons because of its very high critical mass.

Delayed Neutron Fraction: In all reactors, some fraction of the fission neutrons are delayed neutrons emitted from the decay of very-short-lived fission products. This fraction is used for reactor control. Unlike solid fuel reactors, the flowing fuel implies that some fraction of the delayed neutrons will occur in flowing fuel that has left the reactor core. This must be accounted for in all reactor physics calculations and evaluations.

Fuel Cycle Options

Five major fuel cycle options exist to address different goals of reactor operation. The basic reactor remains unchanged except for the salt composition, salt-cleanup systems, ratio of salt-to-moderator ratio, and fuel cycle operations. Any of the MSR/fuel-cycle options can be started up using low-enriched uranium or other fissile materials. With the exception of the breeder reactor fuel cycle, the fuel salt processing for all the other fuel cycles can be performed off-site with removal of the fuel salt every few years.

Actinide Burning: This fuel cycle burns multi-recycle plutonium, americium, and curium from LWR SNF or other sources to reduce the long-term hazard of wastes to an SNF repository. It can also produce denatured ^{233}U as a by-product. The penalty for burning actinides in an epithermal neutron flux is partly offset by the greater fission neutron yield of the higher actinides. As an actinide-burner, the MSRs will produce 10% more electricity than the other reactors that originally generated the actinides. This mode of operation requires a molten salt, such as a sodium-zirconium fluoride salt, that has a high solubility for actinides. In the process of burning actinides, the actinides with high fission cross sections are burnt out first. It requires substantially longer times to burnout low nuclear-cross-section actinides. Consequently, there is a buildup of low cross-section actinides in the reactor. This implies that any reactor burning actinides from LWRs will have a larger inventory of actinides in the reactor core than with other MSR fuel cycles.

Much of the current interest in MSRs is a result of the reactors' capabilities to burn actinides to reduce waste management burdens. Because they are liquid-fuel reactors, MSRs offer three advantages over solid-fuel reactors in this application:

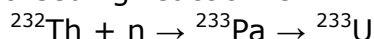
No Isotopic Blending: Different lots of SNF have different Pu, Am, and Cm isotopics. The MSR has a homogeneous liquid fuel. Any fissile material can be fed to the reactor where it mixes with the whole volume of the fuel salt. The different nuclear characteristics of different batches of higher actinides are addressed by the rate of addition to the homogeneous molten fuel salt. In contrast, in solid-fuel reactors, the quantity and isotopics of the fissile materials in every location of every fuel assembly must be controlled to avoid local overpower conditions that burn out the fuel. With complex mixtures of isotopics, the process of mixing fissile materials to obtain uniform solid-fuels is expensive and difficult to accomplish.

No Fuel Fabrication: The higher actinides have small critical masses and high rates of decay heat representing a serious technical and economic challenge for solid-fuel fabrication. This is a non-issue for an MSR because no fuel fabrication is required.

Minimal Reprocessing: In an MSR, fission products are removed from the molten salt, while actinides remain in the salt. This is the reverse of traditional processing, in which clean fissile materials are extracted from SNF. In an MSR, the cleaned fuel salt is to be mixed back with the salt in the reactor. Some fraction of the fission products must be removed, but there is no reason to fully clean the salt. Processing would be done as a batch process at a collocated or off-site location.

Once-Through Fuel Cycle: The once-through fuel cycle converts thorium to ^{233}U internally in the reactor and uses 20% enriched uranium as fresh fuel to the reactor. The annual fuel consumption is ~ 45 t/GWe, or about one-fifth that of an LWR. No recovery of fissile material from the discharged salt would be required.

^{233}U -Thorium Breeder Cycle: MSRs can operate as breeder reactors. After startup, only thorium is added as a fuel. A breeder reactor with efficient fuel production also requires on-line processing of the fuel salt because of the nuclear characteristics of breeding fuel with thermal neutrons using the ^{233}U -thorium fuel cycle. In a thermal neutron breeder reactor, the breeding reaction is



Unfortunately, ^{233}Pa has a moderately large absorption cross section and a half-life of 27 days. If it is left in the reactor, parasitic capture of neutrons by ^{233}Pa will occur resulting in a significant reduction in the breeding ratio. To avoid this scenario and to obtain high breeding ratios, on-line processing is required for removal of the

^{233}Pa and storage outside of the reactor until it decays to ^{233}U . With an efficient processing system, the breeding ratio is ~ 1.06 , with an equilibrium ^{233}U inventory of about 1,500 kg. If the reactor is to be a breeder reactor, the fuel salt characteristics must be optimized and will almost certainly be a mixture of ^7LiF , BeF_2 , ThF_4 , and UF_4 . This salt mixture provides better neutron economy. The use of a ^{233}U -thorium breeder reactor cycle results in a high-level waste with a very low actinide content because as neutrons are added to the thorium, the various fissile isotopes that are produced (^{235}U , ^{239}Pu , etc.) tend to fission.

There has been one important change in the breeder reactor fuel cycles. In the 1960s, it was thought that uranium resources were limited; thus, the goal was to maximize the breeding ratio to provide the fuel for a rapid buildup of additional nuclear power plants. It is now recognized that the uranium resources are much larger than originally estimated. Consequently, existing fissile fuel resources may be sufficient to initially fuel any required number of reactors. In this case, the only long-term requirement is to make fuel as fast as it is consumed. This requires a net breeding ratio of 1. Lowering the required breeding ratio reduces the requirements for the on-line processing of the fuel salt and may allow major simplifications in salt processing. These options are being investigated in Europe.

Denatured ^{233}U -Thorium Breeder Cycle: This is a breeder reactor fuel cycle designed to maximize proliferation resistance by minimal processing of the fuel salt and by addition of ^{238}U to isotopically dilute fissile uranium isotopes. This lowers the breeding ratio to slightly above 1 and results in a very low fissile plutonium (^{239}Pu and ^{241}Pu) inventory of ~ 0.16 kg/MWe. The use of a thorium- ^{233}U breeder reactor cycles results in a high-level waste with a low actinide content because as neutrons are added to the thorium, the various fissile isotopes that are produced (^{235}U , ^{239}Pu , etc.) tend to fission.

Fast Spectrum Uranium-Plutonium Breeder Cycle: The improved understanding of salt chemistry has resulted in the development of MSR concepts that use salts that allow very high loadings of fissile materials. The high fissile loadings create the option to have a fast spectrum MSR. This enables the use of the same fuel cycle options as with sodium-, helium-, and lead-cooled fast reactors.

Scope of Research and Development

A credible understanding of the economics, the nature of application (electricity generation, hydrogen production, actinide burning, or fissile fuel production) decide the basis for a decision on whether to initiate a large-scale developmental program with the goal of deployment.

Design Optimization: Three major changes need to be incorporated into a modern MSR design as compared to the MSBR design of 1970s by ORNL. First, the new

high-temperature technologies (such as Brayton cycles) must be incorporated into the MSR design because they simultaneously eliminate previously identified technical challenges associated with earlier MSR designs, improve plant efficiency, and reduce the capital cost per kWe. Second, advances in remote operations, robotics, and controls must be incorporated into the conceptual design. Last, changing application requirements will simplify plant design. Early MSRs were designed to maximize fuel production; that mission, in turn, required complex, high-capacity, on-line salt processing. For actinide burner and hydrogen missions, there is the potential to eliminate most on-line fuel processing systems and greatly simplify the plant design.

Regulation: Liquid fueled reactors use different approaches to reactor safety than solid fueled reactors. These include: (1) draining the fuel into critically safe, passively cooled tanks if off-normal conditions occur, (2) limiting excess reactivity by online fuel processing and/or continuous fueling, and (3) limiting fission product source terms by on-line processing. The current regulatory structure was developed with the concept of solid-fuel reactors. The comparable regulatory requirements for this system must be defined. Using current tools, appropriate safety analysis is required followed by appropriate research on the key safety issues.

Safety: The critical safety requirement for an MSR is that the radionuclides remain dissolved in the molten salt under all conditions. The reactor size, design, and safety systems are dependent upon this property. There are two basic R&D tasks: (1) determine the limits of the solubility of trivalent actinides in candidate molten salts and (2) assure control of noble metal fission products in the primary system. New applications for MSRs, such as actinide burning, imply higher concentrations of trivalent actinides and noble metals in the salt than were used in the past and may require modification of the salt composition to assure solubility under all conditions. R&D is required to determine the trivalent solubility limits under these different conditions. Similarly, the behavior of noble metal fission products in the salt and their ultimate disposition is required. Under some conditions, fission product noble metals may plate out on heat exchangers resulting in high decay heat loads and limited equipment lifetimes.

Fuels and Fuel Cycles: There are multiple fuel cycle challenges. Some are common to other reactors and their associated fuel cycles, and some are unique to the MSR. Specifically, because the system is a molten fluoride salt system, there are unique chemical issues not associated with other reactors. There is a need to develop a fluoride high-level waste form and an integrated fuel recycle strategy. Since earlier MSR efforts, there have been major advances in separation technologies and proposals for highly innovative separation systems unique to fluoride salts.

Energy Conversion: The goal of the energy conversion R&D is to establish the technical basis for coupling Brayton cycles for electricity production and thermochemical water cracking cycles for hydrogen production to MSRs. These activities are expected to take place as part of an effort on crosscutting energy conversion R&D.

Materials: The major goal of the materials R&D is to identify and qualify materials with properties appropriate for MSR operating conditions, including corrosion resistance, mechanical performance, and radiation performance. The primary materials of interest are the moderator (graphite) for thermal neutron spectrum MSRs and the reactor vessel/primary loop alloy (presently a Ni-based alloy). It is also necessary to develop corrosion control and coolant monitoring strategies for protecting the reactor vessel and primary piping alloys. There is also a natural base to build on to extend candidate materials for the higher temperature regimes for realizing higher efficiency. Mention can also be made of use of graphite as a moderator and various carbon-carbon composites for multiple structural applications, and Ni-based super alloys and other alloys for reactor components.

Introduction

Preamble

In the early days of nuclear energy, especially in the USA as a follow-up of the Manhattan Project (1945–60), the leading US laboratories singled out thorium as a possible complement for uranium and the possibility of using U-233 as a nuclear weapon. In 1958, about 55 kg of U-233 were available in the US. In the Atoms for Peace Programme, with its great variety of developments (1955–75), thorium appeared to be an interesting resource for supplementing limited uranium reserves in the context of a fast-developing nuclear industry. During that pioneer period, about 1500 kg of U-233 have been separated in the USA from 900 tonnes of thorium. The US and France have each separated about 2000 tonnes of thorium, part of which is still available. A flurry of pre-industrial projects dealing with demonstration reactors and study of the thorium fuel cycle were initiated. Many reactor prototypes were built and operated. Mention may be made of:

PEACH BOTTOM (40 MWe) of General Atomics, has run successfully from 1966 to 1972

The DRAGON experimental reactor (20MWt; 1966–73) was built and operated successfully as an international OECD–EURATOM venture in England, also involving Sweden, Norway and Switzerland.

The German experimental pebble-bed AVR in Julich (15 MWe) built with BBC-HRB and operated between 1967 and 1989, has been a very remarkable and quite exceptional reactor due to its revolutionary design and excellent performance.

Following these encouraging results, General Atomics and HRB each built an industrial prototype of 300 MW, Fort St Vrain (1976–89) and THTR (1985–89) extrapolated from PEACHBOTTOM and AVR respectively.

Following the success of the AVR, Siemens developed the modular HTR concept, called MODUL. The idea was to combine small HTR-modules (200 MWth) to power stations of adjustable output.

History of molten salt reactors

Two experimental reactors were built and successfully operated. The Aircraft Reactor Experiment (ARE) was the first MSR. It was a 2.5-MW(t) reactor that was operated in 1954 at a peak temperature of 860°C and used a sodium-zirconium fluoride salt. This was followed in 1965 by the Molten Salt Reactor Experiment (MSRE), an 8-MW(t) reactor. This reactor, based on a lithium, beryllium and zirconium fluoride salt was operated with 30% ^{235}U enriched uranium from 1965 to 1968, with ^{233}U from 1968 to 1969 and finally with plutonium mixed with ^{233}U in 1969 and demonstrated most of the key technologies for a power reactor. These two

experimental MSRs built at Oak Ridge National Laboratory (ORNL) established the basic technology for the MSR and their success warranted the study of a thorium power breeder reactor coupled to a reprocessing unit for the on-line extraction of fission products: the Molten Salt Breeder Reactor (MSBR). In addition, test loops with molten salts were operated for hundreds of thousands of hours, materials of construction were code qualified to 750°C, and a detailed conceptual design of a 1000-MW(e) MSBR was developed. Over a 1000 technical reports were produced.

After 1975, the Ford–Carter policy in the US, the success of the light-water reactors, a slower development of nuclear energy and other factors, brought the thorium projects to a halt, except in India.

It is interesting to recall the predictions of the International Fuel Cycle Evaluation Conference (INFCE) of 1978 (published in 1980 by IAEA), where thorium was given almost equal importance as uranium. It appeared that if the optimistic nuclear energy predictions were to be followed, thorium was to be called upon massively in the future. These predictions were too optimistic, but in the very long-term, the use of thorium along with uranium could increase the potential of nuclear energy considerably. Studies on this sort of reactor were therefore resumed in the 1980's in Japan with the THORIMS-NES then the FUJI-AMSB, as well as in France with studies on the MSBR by EDF and the CEA. In the 1990's, the concept was taken up again with a view to incinerating nuclear wastes in sub-critical reactors such as the TIER-1 project, to transmute the plutonium of pressurized water reactors, the CEA TASSE project, or the EDF AMSTER project.

It should be mentioned that the monumental and seminal research work and contributions from ORNL on the molten salt reactor concept is still the forerunner of all the subsequent models and copious open literature available on the ORNL work has been freely adopted. The two molten salt reactor projects at ORNL established the basic technologies for the MSR and demonstrated its main advantages namely good neutron economy, inherent safety, on-line refueling, and processing and fission product removal. Due to these advantages, the MSR has been included as one of six Generation IV reactor concepts. Some new MSR concepts include: small molten salt reactor (SMSR), the actinides molten salt transmuter (AMSTER), MOST project and MOSART and thorium molten salt reactor (TMSR). Since 2005, R&D has focused on the development of fast-spectrum MSR concepts (MSFR) combining the generic assets of fast neutron reactors (extended resource utilization, actinide burning, waste minimization) to those relating to molten salt fluorides as fluid fuel and coolant (favourable thermal-hydraulic properties, high boiling temperature, optical transparency). In addition, MSFR exhibit large negative temperature and void reactivity coefficients, a unique safety characteristic not found in solid-fuel fast reactors. MSFR has been recognized as a long term alternative to solid-fuelled fast neutron systems with unique potential (negative feedback coefficients, smaller fissile inventory, easy in-service inspection, simplified fuel cycle...).

Apart from MSR systems, other advanced reactor concepts are being studied, which use the liquid salt technology, as a primary coolant for the Advanced High-Temperature Reactor (AHTR) or intermediate coolant, as an alternative to secondary sodium, for Sodium Fast Reactors (SFR) and to intermediate helium for Very High Temperature Reactors (VHTR).

The renewal and diversification of interests in molten salts have led the MSR Provisional System Steering Committee (PSSC) to a shift of the R&D orientations and objectives and two baseline concepts are considered which have large commonalities in basic R&D areas, particularly for liquid salt technology and materials behavior (mechanical integrity, corrosion):

- The Molten Salt Fast neutron Reactor (MSFR) is a long-term alternative to solid-fuelled fast neutron reactors offering very negative feedback coefficients and simplified fuel cycle. The potential of MSFR has been assessed but specific technological challenges must be addressed and the safety approach has to be established.
- The Advanced High Temperature Reactor (AHTR) is a high temperature reactor with higher power density than the VHTR and passive safety potential from small to very high unit power (> 2 400 MWt).

In Russia, the efficiency of MSR for actinide burning has been investigated. This resulted into the single stream Li, Na,Be/F MOlten Salt Actinide Recycler & Transmuter (MOSART) fast spectrum system fuelled with compositions of plutonium plus minor actinide trifluorides (AnF₃) from UOX and MOX LWR spent fuel without U-Th support.

In addition, the opportunities offered by liquid salts for intermediate heat transport in other systems (SFR, VHTR) are being investigated. Liquid salts offer two potential advantages: smaller equipment size because of the higher volumetric heat capacity of the salts; and no gross chemical exothermal reactions between the reactor, intermediate loop, and power cycle coolants.

Liquid salt chemistry plays a major role in the viability demonstration of MSR and AHTR concepts with such essential R&D issues as: (a) the physico-chemical behaviour of coolant and fuel salts, including fission products and tritium, (b) the compatibility of salts with structural materials for fuel and coolant circuits, as well as fuel processing materials development, (c) the on-site fuel processing, (d) the maintenance, instrumentation and control of liquid salt chemistry (redox, purification, homogeneity), and (e) safety aspects, including interaction of liquid salts with sodium, water, and air.

MSFR REFERENCE OPTIONS

The primary feature of the MSFR (Molten Salt Fast Reactor) concept is the removal of the graphite moderator from the core (graphite-free core). In terms of fuel

cycle, two basic options have been investigated, ^{233}U -started MSFR and TRU-started MSFR. Realistic drawings showing the main MSFR components and their arrangement in the vessel have been elaborated. Figure 1 displays a schematic drawing of a vertical section of the MSFR while Table 1 presents some characteristics of the reactor. The core is a single cylinder (diameter equal to height) where nuclear reactions take place within the flowing fuel salt. It is made of three volumes: the active core, the upper plenum and the lower plenum. The fuel salt is a binary salt, composed of LiF enriched in ^7Li (99.999%) and heavy nuclei (HN) amongst which the fissile element, ^{233}U or Pu. The $(\text{HN})\text{F}_4$ proportion is set at 22.5 mol% (eutectic point), corresponding to a melting temperature of 550°C . The choice of this fuel salt composition relies on many systematic studies (influence of chemical reprocessing on neutronic behavior, burning capabilities, deterministic safety level, and deployment capabilities). This salt composition leads to a fast neutron spectrum in the core. The outer core structures and heat exchangers are protected by thick reflectors designed to absorb more than 80% of the escaping neutron flux. These reflectors are themselves surrounded by a 10 cm thick neutronic protection of B_4C absorbing remaining neutrons. Axial reflectors are made of nickel-based alloys. The radial reflector consists of a fertile blanket (50 cm thick) filled with a fertile salt of LiF-ThF₄ with 22.5 mol% ^{232}Th .

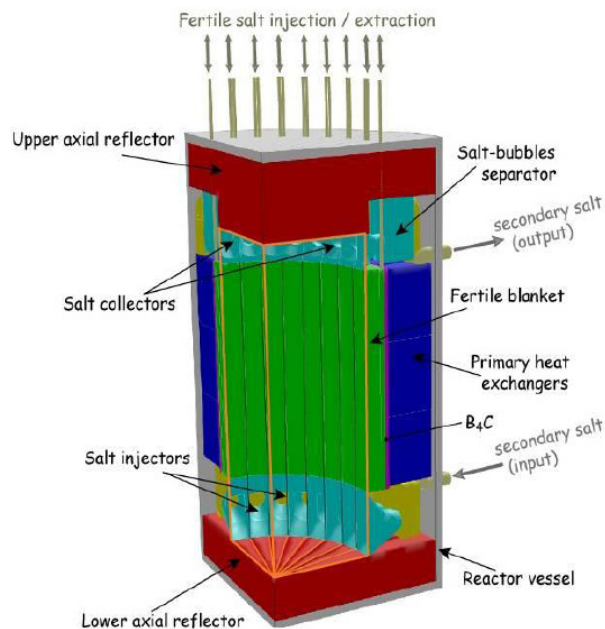


Figure 1: Schematic view of a quarter of the MSFR

The level of deterministic safety reached by the concept is excellent since the feedback coefficients of the MSFR are negative in both ^{233}U and TRU starting modes. The total feedback coefficient is equal to $-6 \text{ pcm}/^\circ\text{C}$ when the equilibrium state of the reactor has been reached and the density coefficient, which for MSRs can also be viewed as a void coefficient, is also largely negative at about $-3 \text{ pcm}/^\circ\text{C}$.

A good indicator of the deployment capability is the doubling time, defined by the operation time leading to the ^{233}U inventory of a new reactor of the same type through breeding. For a ^{233}U -MSFR, the annual ^{233}U production is 120 kg which corresponds to 50 years doubling time per reactor. Starting a MSFR from Generation II or III reactors spent fuel is more favourable and yields 35 years doubling time. Indeed, the presence of other fissile elements PB-AHTR core section decreases the consumption of ^{233}U and improves the deployment capability of the concept.

Table 1: Reference design characteristics of the MSFR

Thermal power (MWt)	3000				
Fuel molten salt composition (mol%)	LiF-ThF ₄ - ²³³ UF ₄ or LiF-ThF ₄ -(Pu-MA)F ₃ with LiF = 77.5 mol%				
Fertile blanket molten salt composition (mol%)	LiF-ThF ₄ (77.5-22.5)				
Melting point (°C)	550				
Operating temperature (°C)	700-800				
Initial inventory (kg)	²³³ U-started MSFR		TRU-started MSFR		
	Th	²³³ U	Th	Actinide	
	38300	5060	30600	Pu	11200
				Np	800
				Am	680
Cm				115	
Density (g/cm ³)	4.1				
Dilatation coefficient (/°C)	10 ⁻³				
Core dimensions (m)	Radius: 1.15 Height: 2.30				
Fuel salt volume (m ³)	18 9 out of the core 9 in the core				
Blanket salt volume (m ³)	8				
Thorium consumption (ton/year)	1.112				
²³³ U production (kg/year)	93 (²³³ U-started MSFR) 188 during 20 years then 93 (TRU-started MSFR)				
Breeding ratio (²³³ U-started MSFR)	1.085				

AHTR Reference Plant Concept

The AHTR is a new reactor concept that has three technical characteristics: high temperature, passive safety, and a large power output. The high temperature is

required to produce hydrogen and efficiently produce electricity. The passive safety features are required to reduce operating costs and improve public acceptance. The large power output, passive safety features, and high efficiency of electricity production (a consequence of high temperatures) are the enabling technologies to improve economics. The development is a joint effort of Oak Ridge National Laboratory, Sandia National Laboratories, and the University of California at Berkeley.

The defining aspects of an Advanced High Temperature Reactor (AHTR) are the use of coated particle fuel embedded within a graphitic matrix cooled by liquid fluoride salt. A Pebble Bed Advanced High Temperature Reactor (PB-AHTR) operating at ~900 MWt is the most actively developing commercial scale plant design. The plant design is currently transitioning from a primarily conceptual to an initial engineering scoping phase. A half cross section of the core concept is shown in Figure 2.

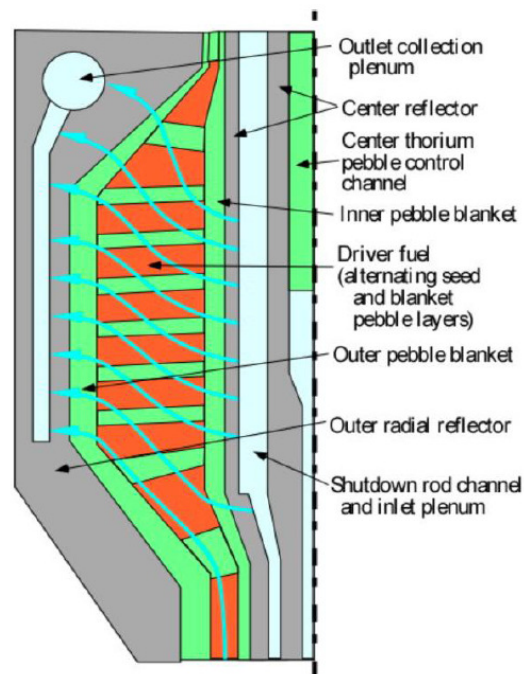


Figure 2: Half Cross Section of PB-AHTR Core

A major design refinement of the current core is the use of inner and outer pebble blankets to reduce the radiation damage to the fixed reflector graphite. The power density of a salt cooled pebble bed is 4-8x greater than that of its gas-cooled cousin. The resultant higher flux level would necessitate more frequent reflector graphite replacement without the use of blanket pebble layer. The controlled motion of a structured pebble assembly has recently been demonstrated using simulant materials at U.C. Berkeley, along with friction coefficient measurements for graphite pebbles verifying that fluoride salts act as effective lubricants and that friction coefficients are very close to those for the simulant materials. Pebble motion demonstration using

prototypic materials and temperatures will be a key aspect of future R&D on the PB-AHTR.

R&D PROGRESS AND REMAINING ISSUES IN SPECIFIC AREAS

Significant progress has been achieved in 2008 in critical areas of MSR-AHTR R&D. In brief, the essential facts are the following:

1. Salt selection for different applications is stabilized, the needs of complementary data have been clarified.
2. A strongly improved (versus MSBR) fuel salt clean-up scheme has been developed.
3. Criticality tests are being performed for the assessment of MSR and AHTR fuel and core behaviour.
4. Although progress has been made in the area, the assessment of structural materials remains challenging for MSFR and AHTR as both concepts are supposed to operate at temperatures higher compared to MSBR.

Salt selection for different applications

The ${}^7\text{LiF}\text{-BeF}_2$ (66:34 in mol%) salt is the selected fuel carrier for the moderated (thermal) molten salt thorium breeder, giving as fuel salt ${}^7\text{LiF}\text{-BeF}_2\text{-ThF}_4\text{-UF}_4$. From neutronic as well as chemical point of view, there are no alternatives for this salt that do not penalise the breeding capacity of the reactor.

${}^7\text{LiF}\text{-ThF}_4$ (78:22 or even 71:29 in mol%) is the reference fuel solvent composition for the fast spectrum molten salt thorium breeder reactor (MSFR). The neutronic analysis of the MSFR concept has demonstrated the feasibility of the concept, but it must still be clarified whether the physico-chemical properties (melting temperatures, solubility for the actinide trifluorides, density, expansivity, viscosity, thermal conductivity, heat capacity) of this salt fuelled by significant amount of UF_4 (2-4% of the total heavy nuclei in the moderated and 12-18% in the fast systems) or AnF_3 (up to 25% of the total heavy nuclei in the fast concept) are consistent with safe operation of the reactor and fuel salt clean-up unit. To tune these properties, addition of other components is possible. The most obvious is BeF_2 but there is an incentive to keep the content of this material low (e.g. 71LiF-2BeF₂-27ThF₄ or 75LiF-5BeF₂-20ThF₄ in mol%) or even zero. Alternatives are NaF and possibly CaF₂. Therefore, the ${}^7\text{LiF}\text{-NaF}\text{-ThF}_4$ system must be further analysed, whereas scoping studies of the ${}^7\text{LiF}\text{-CaF}_2\text{-ThF}_4$ system are required, to assess the pros and cons for both molten salt mixtures, including suitability for fuel salt processing.

The molten salt actinide burner is a fast spectrum concept too. The carrier salt for this application must have good solubility for the actinide trifluorides and this can be achieved using ${}^7\text{LiF}\text{-NaF}\text{-(KF)}$ as solvent or ${}^7\text{LiF}\text{-(NaF)}\text{-BeF}_2$ melt. Again, the goal is to keep the content of BeF_2 low or even zero. An interesting alternative is the use of plutonium and minor actinides as start-up for the thorium cycle in the MSR, leading to ${}^7\text{LiF}\text{-NaF}\text{-ThF}_4$ carrier salt.

In summary, it is clear that the ${}^7\text{LiF-(NaF)-AnF}_4\text{-AnF}_3$ salt (where An represent actinides) is the key system to be further investigated in parallel to the ${}^7\text{LiF-(NaF)-BeF}_2\text{-AnF}_4\text{-AnF}_3$ system. Optimisation of the fractions of the components is still needed with respect to mentioned-above physico-chemical properties, corrosion behavior in the Ni-Mo alloys and fuel salt processing.

Table 2: Fuel and coolant salts for different applications

Reactor type	Neutron spectrum	Application	Carrier salt	Fuel system
MSR-Breeder	Thermal	Fuel	${}^7\text{LiF-BeF}_2$	${}^7\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$
	Non-moderated	Fuel	${}^7\text{LiF-ThF}_4$	${}^7\text{LiF-ThF}_4\text{-UF}_4$
				${}^7\text{LiF-ThF}_4\text{-PuF}_3$
MSR-Breeder	T/NM	Secondary coolant	NaF-NaBF_4	
MSR-Burner	Fast	Fuel	LiF-NaF	$\text{LiF-(NaF)-AnF}_4\text{-AnF}_3$
			LiF-(NaF)-BeF_2	$\text{LiF-(NaF)-BeF}_2\text{-AnF}_4\text{-AnF}_3$
			LiF-NaF-ThF_4	
AHTR	Thermal	Primary coolant	${}^7\text{LiF-BeF}_2$	
SFR		Intermediate coolant	$\text{NaNO}_3\text{-KNO}_3\text{-(NaNO}_2\text{)}$	

For coolant salts, one has to make a distinction between salt for in-core use (primary coolant) and salts for out-of-core use (secondary or intermediate coolants). For primary coolants in thermal reactors, the requirements are very similar to thermal breeder reactors and ${}^7\text{LiF-BeF}_2$ (66-34 with $T_m=458^\circ\text{C}$) is the main candidate, with ${}^7\text{LiF-NaF-KF}$ (46-11.5-42.5 with $T_m=454^\circ\text{C}$), LiF-NaF-RbF (46.5-6.5-47 with $T_m=426^\circ\text{C}$) and ${}^7\text{LiF-NaF-BeF}_2$ (30.5-31-38.5 with $T_m=316^\circ\text{C}$) as alternatives. Note that the last alternative molten salt mixture has the lowest liquidus temperature.

For secondary coolant applications, neither neutronic considerations nor actinide solubility play a role and a wider choice of materials is possible. For MSRs in which tritium control is the main concern, the NaF-NaBF_4 (8:92 with $T_m=385^\circ\text{C}$) system is the prime candidate, mainly because of its satisfactory tritium trapping. A ternary salt LiF-NaF-BeF_2 should be considered in future studies as alternative secondary coolant because a freezing temperature range of about $315\text{-}335^\circ\text{C}$ would be a practical value for engineering consideration. Because closed gas Brayton cycles can mitigate both the tritium and the melting point concerns, LiF-NaF-KF or NaCl-KCl-MgCl_2 may also be considered as a secondary salt.

Finally, heat transfer for lower temperature applications (below 600°C) requires a cheap and stable salt. $\text{NaNO}_3\text{-KNO}_3$ possibly with addition of NaNO_2 is the main candidate identified at this stage.

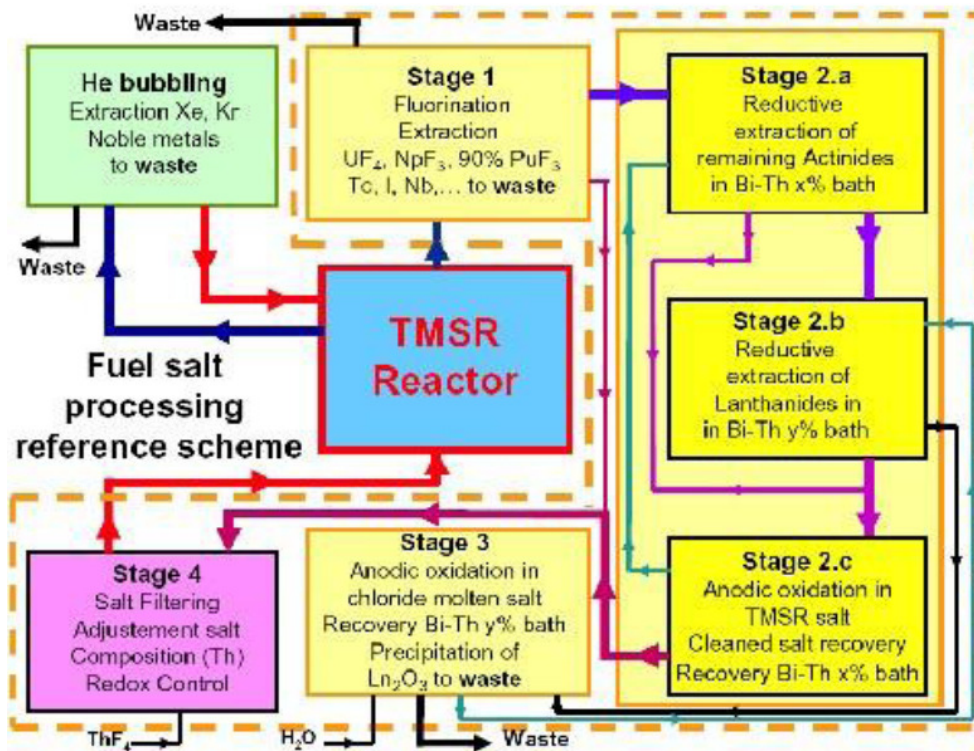


Figure 3: TMSR (MSFR) reference fuel salt processing

Fuel salt clean-up scheme

The salt processing scheme relies on both on-line and batch processes to satisfy the constraints for a smooth reactor operation while minimizing losses to waste streams. ORNL experiments have provided some data mainly for the on-line gaseous fission product extraction process.

Acquisition of fundamental data for the separation processes is needed especially for the actinide-lanthanide separation. The extraction of lanthanides has to be done because of the low solubility of these trifluoride elements and neutronic captures that decrease the reactivity balance.

The progress made in core design in the last two years has opened the door for the definition of an improved fuel salt reprocessing scheme with a realistic fuel clean-up rate (40 l/day) and minimized losses to wastes.

The proposed reference processing scheme is shown in Figure 3. The first step (green box) involves an on-line gaseous extraction with helium bubbling to remove gaseous fission products, Xe and Kr, and a part of the noble metals from fuel circuit. On the other hand, a batch fuel process separates the actinides which are returned to the reactor salt from the harmful fission products (mostly lanthanides). The fuel

clean-up rate has been set at 40 liters per day, corresponding to the processing of 100 kg heavy nuclei per day. This value is almost two orders of magnitude less than the reference MSBR scheme.

The reference scheme depicted in Figure 3 involves 4 stages for the batch on-site fuel processing. The peculiarity of the concept appears in stages 2 and 3 by combining chemical and electrochemical methods for the extraction and the back extraction of actinides and lanthanides. This choice leads to fuel processing without effluent volume variation and the fuel processing balance is reduced to only one reaction: $2\text{LnF}_3 + 3\text{H}_2\text{O}(\text{g}) = \text{Ln}_2\text{O}_3 + 6\text{HF}(\text{g})$.

Critical steps of the new fuel clean-up scheme are addressed and will be experimentally assessed in new facilities. The design and construction of a molten salt loop to study both He bubbling efficiency and material corrosion attack has been initiated. An efficient technique for actinide/lanthanide separation is under qualification.

Criticality tests for the assessment of MSR and AHTR fuel and core behaviour

The SPHINX (SPent Hot fuel Incinerator by Neutron flux) project was originally defined as a suitable experimental basis at representative scale for the demonstration of MSR-burner feasibility. It relies on the utilization of the zero power experimental reactor LR-0 being operated in the Nuclear Research Institute Rež (NRI), Czech Republic. This full-scale physical model of the PWR cores was modified in order to allow the measurement of all the neutronic characteristics of the MSR burner and/or breeder blanket, at first by room temperature and in future stage by conditions close to operational. (Figure 4).



Figure 4: LR-0 zero power critical test facility

Because two baseline concepts (MSFR, AHTR) are now considered, a corresponding broadening of the SPHINX project was discussed and formally adopted at the end of 2008. The LR-0 will thus be used for the validation of AHTR neutronics models (reactivity coefficients...) in the frame of a collaboration between the Czech Republic (NRI) and USA (University of California, Berkeley).

Two versions of EROS elementary blocks, as simplified models of the AHTR core module, have been designed and manufactured. During December 2008, the critical tests of both those elementary blocks were performed. The simplified models are completely ready for complex testing of experimental and measuring methods for detailed neutron field distribution and principle neutronic characteristics prediction.

AHTR with VIPAC fuel

The AHTR (Figure 5 Table 3) uses coated-particle graphite-matrix fuels and a molten-fluoride-salt coolant. The fuel is the same type that is used in modular high-temperature gas-cooled reactors (MHTGRs), with fuel failure temperatures in excess of 1600°C. The optically transparent molten salt coolant is a mixture of fluoride salts with freezing points near 400°C and atmospheric boiling points of ~1400°C. Several different salts are being evaluated as the primary coolant, including lithium-beryllium and sodium-zirconium fluoride salts. The reactor operates at near-atmospheric pressure. At operating conditions, the molten-salt heat-transfer properties are similar to those of water. Heat is transferred from the reactor core by the primary molten-salt coolant to an intermediate heat-transfer loop. The intermediate heat-transfer loop uses a secondary molten-salt coolant to move the heat to the turbine hall. In the turbine hall, the heat is transferred to a multi-reheat nitrogen or helium Brayton cycle power conversion system. For hydrogen production, the heat is transferred to the thermochemical hydrogen production facility, which converts water and high temperature heat to hydrogen and oxygen.

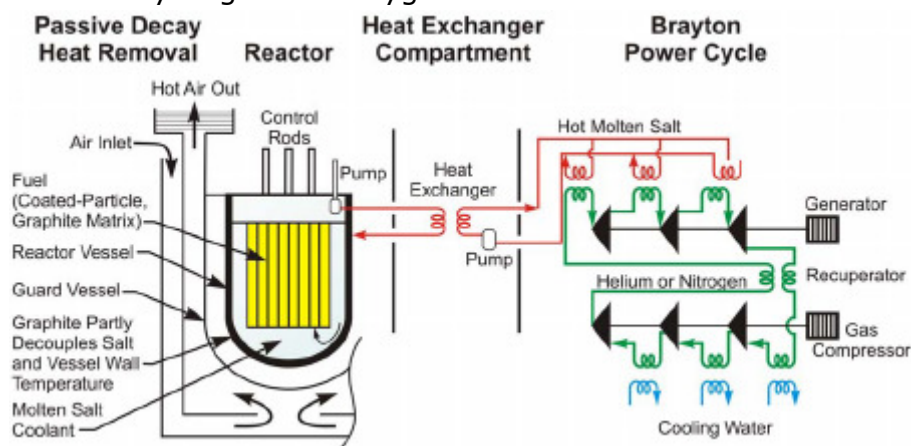


Figure 5 Schematic of the AHTR for electricity production

Table 3. AHTR preconceptual design parameters

Power level	2400 MW(t)	Power cycle	3-stage multi-reheat Brayton
Core inlet/outlet temperatures (options)	900 ⁰ C/1000 ⁰ C 700 ⁰ C/800 ⁰ C 670 ⁰ C/705 ⁰ C	Electricity (output at different peak coolant temperatures)	1357 MW(e) at 1000 ⁰ C 1235 MW(e) at 800 ⁰ C 1151 MW(e) at 705 ⁰ C
Coolant	2 ⁷ LiF-BeF ₂ (NaF-ZrF ₄)	Power cycle working fluid	Nitrogen (Helium: long term option)
Fuel		Vessel	
Kernel	U-carbide/oxide	Diameter	9.2 m
Enrichment	10.36 wt% ²³⁵ U	Height	19.5 m
Form	Prismatic	Reactor core	
Block diameter	0.36 m (across flats)	Shape	Annular
Block height	0.79 m	Diameter	7.8 m
Columns	324	Height	7.9 m
Decay heat system	Air cooled	Fuel annulus	2.3 m
Volumetric flow rate	5.54 m ³ /s	Power density	8.3 W/cm ³
Coolant velocity	2.32 m/s	Reflector (outer)	138 fuel columns
		Reflector (inner)	55 fuel columns

The baseline AHTR facility layout (Figure 6) that was developed is similar to the S-PRISM sodium-cooled fast reactor designed by General Electric. Both reactors operate at low pressure and high temperature; thus, they have similar design constraints. The 9.2-m-diam. vessel is the same size as that used by the S-PRISM. In the initial baseline studies, it was assumed that the fuel and power density (8.3 W/cm³) were essentially identical to those of the MHTGR. The better heat transfer of the molten salt relative to helium implies (1) lower peak reactor temperatures if the power densities are similar or (2) a higher-power-density core that is smaller or has a higher total power output for a given reactor vessel size. Three peak coolant temperatures are being considered: 705, 800, and 1000°C for the AHTR—Low Temperature (AHTR-LT), the AHTR—Intermediate Temperature (AHTR-IT), and the AHTR—High Temperature [AHTR-HT], respectively. The AHTR-LT uses existing materials, the AHTR-IT uses existing materials that have not been fully tested, and the AHTR-HT uses advanced materials. The AHTR-HT and AHTR-IT include a graphite blanket system, while the AHTR-LT has a metallic blanket system that separates and insulates the reactor vessel from the reactor core so that the fuel and coolant can operate at higher temperatures than the vessel. This insulation ensures long vessel life (minimizing long term creep) and minimizes heat losses during normal operations.

In the proposed design, the AHTR has an annular core through which the coolant flows downward. The molten salt coolant flows upward through the non-fuel graphite section in the middle of the reactor. The molten-salt-coolant pumps and their

intakes are located above the reactor core with appropriate siphon breakers; thus, the reactor cannot lose its coolant except by failure of the primary vessel. The guard vessel is sized so that even if the primary vessel fails, the core remains covered with salt.

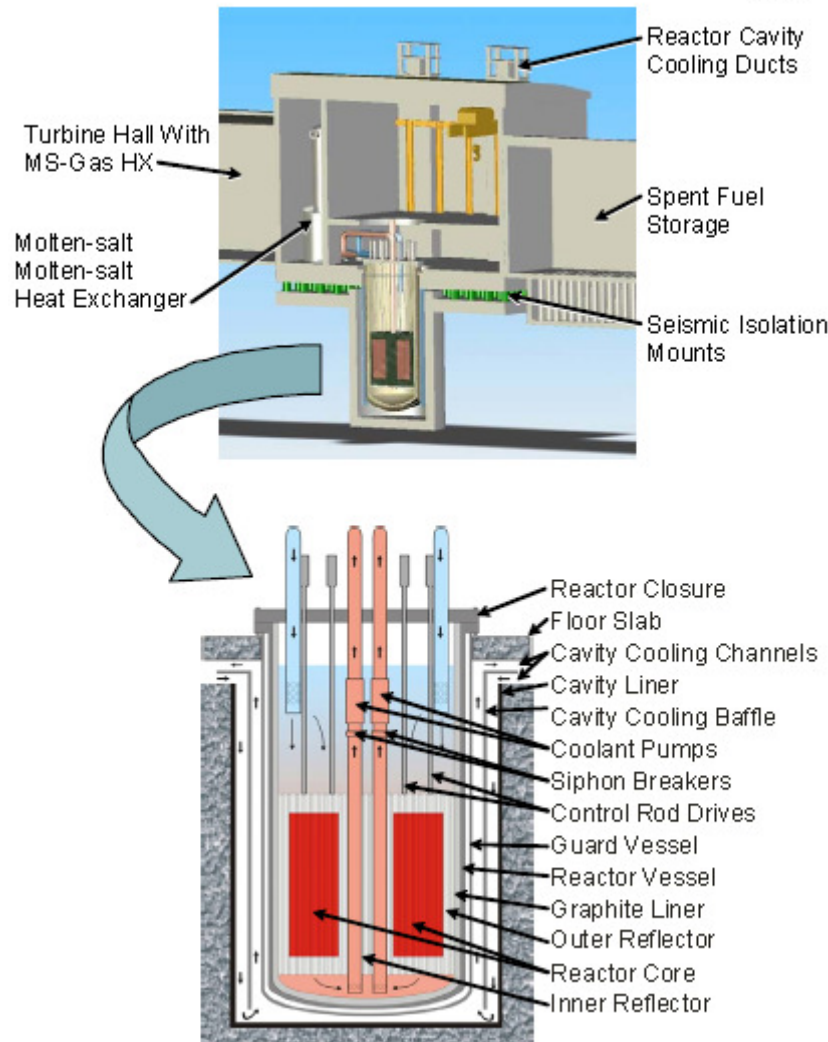


Figure 2 Schematic of the AHTR nuclear island and vessel

Decay Heat Cooling and Accident Management

When a reactor shuts down, radioactive decay heat continues to be generated in the reactor core at a rate that decreases over time. The AHTR uses passive reactor vessel auxiliary cooling (RVAC) systems similar to that developed for decay heat removal in the General Electric sodium-cooled S-PRISM. The reactor and decay-heat-cooling system are located in a below-grade silo (Figure 1). In this pool reactor, RVAC system decay heat is (1) transferred from the reactor core to the reactor vessel graphite reflector by natural circulation of the molten salts, (2) conducted through the

graphite reflector and reactor vessel wall, (3) transferred across an argon gap by radiation to a guard vessel, (4) conducted through the guard vessel, and then (5) removed from outside of the guard vessel by natural circulation of ambient air.

The rate of heat removal is controlled primarily by the radiative heat transfer through the argon gas from the reactor vessel to the guard vessel. Radiative heat transfer increases by the temperature to the fourth power (T^4); thus, a small rise in the reactor vessel temperature (as would occur upon the loss of normal decay-heat-removal systems) greatly increases heat transfer out of the system. The effective thermal inertia, per unit volume of the reactor vessel, is much larger than that for gas-cooled reactors because of the high heat capacity of the molten salt and the low radial temperature gradient across the reactor core.

Under accident conditions such as a loss-of-forced-cooling accident, natural circulation flow of molten salt up the hot fuel channels in the core and down by the edge of the core rapidly results in a nearly isothermal core with about a 50°C temperature difference between the top and bottom plenums. For a typical simulation of the reactor with a nominal coolant exit temperature of 1000°C, the calculated peak fuel temperature in such an accident is ~1160°C, which will occur at ~30 hours with a peak reactor vessel temperature of ~750°C at ~45 hours. The average core temperature rises to approximately the same temperature as the hottest fuel during normal operations.

In terms of passive decay-heat-removal systems, a major difference is noted between the liquid-cooled AHTR and gas-cooled reactors. The AHTR can be built in very large sizes [>2400 MW(t)], while the maximum size of a gas-cooled reactor with passive-decay-heat removal systems is limited to ~600 MW(t). The controlling factor in decay heat removal is the ability to transport this heat from the center of the reactor core to the vessel wall or to a heat exchanger in the reactor vessel. The AHTR uses a liquid coolant where natural circulation can move very large quantities of decay heat to the vessel wall with a small coolant temperature difference (~50°C). Unfortunately, in a gas-cooled reactor under accident conditions when the reactor is depressurized, the natural circulation of gases is not very efficient to transport heat from the fuel in the center of the reactor to the reactor vessel. The heat must be conducted through the reactor fuel to the vessel wall. This inefficient heat transport process limits the size of the reactor to ~600 MW(t) to ensure that the fuel in the hottest location in the reactor core does not overheat and fail.

In terms of various accidents, there are no major stored energy sources within containment such as high pressure fluids [helium and water] or reactive fluids [sodium]. This reduces requirements for the containment. The molten salt dissolves almost all actinides and fission products (except xenon and krypton). This provides a major barrier against the release of any radionuclide from fuel failure.

Reactor Physics and Fuel Cycle

Because the AHTR uses the same basic fuel type and the molten salt coolant has a low neutron-absorption cross section, the reactor core physics and fuel cycle options are generally similar to those for helium cooled high-temperature reactors. Reactor power is limited by a negative temperature coefficient, control rods, and other emergency shutdown systems.

Several molten fluoride salts with generally similar properties are being evaluated to determine the optimum coolant salt (Ingersoll et al. 2004). Such evaluations involve trade-offs in neutronics, cost, operations, and other parameters. The initial baseline AHTR design used the same salt ($^{27}\text{LiF}-\text{BeF}_2$) that was used in the MSRE. This salt is well understood and has a negative coolant void coefficient. Several other salts, such as the sodium-zirconium fluoride salts, have operational and cost advantages. Conceptual core design studies with more heterogeneous core designs are under way to determine if negative coolant void coefficients can be obtained for a wide variety of fluoride salts.

Economics

As shown in Table 2, preliminary overnight capital costs of the AHTR for several exit temperatures were determined relative to other higher-temperature reactor concepts [i.e., the S-PRISM and the gas turbine–modular helium reactor (GT-MHR)] based on the relative size of systems and quantities of materials. *This approach provides relative, but not absolute, costs.* Only the construction of multiple reactors provides reliable absolute costs. The lower capital costs are a consequence of several factors: economics of scale [2400-MW(t) reactor vs a 600-MW(t) or 1000-MW(t) reactor, passive safety in a large reactor system, and higher thermal efficiency.

Table 2 Comparison of estimated overnight capital cost (2002\$) of the AHTR-IT and AHTR-HT, as a percentage of the costs of the S-PRISM and GT-MHR [with multi-module output of 1145 MW(e)]^a

	S-PRISM \$1681/kW(e)	GT-MHR \$1528 /kW(e)
AHTR-IT \$930/kW(e)	55%	61%
AHTR-HT \$816/kW(e)	49%	53%

^a The General Electric S-PRISM consists of four reactor modules, each producing 1000 MW(t) and 380 MW(e). The peak sodium temperature is 510°C. The General Atomics GT-MHR consists of four reactor modules, each producing 600 MW(t) and 285 MW(e). The peak helium temperature is 850°C.

Research and Development (R&D)

About 80% of the R&D required for the AHTR is shared with that for helium-cooled high-temperature reactors. This includes fuel development, materials

development, and Brayton power cycles. Many other areas are in common with facility design for liquid-metal reactors. The required R&D is sensitive to the peak coolant temperatures. Near 700°C, existing materials and fuels may be used. At 1000°C, major material development programs are required. Other areas that require significant R&D include (1) the reactor vessel insulation system, (2) optimization of core design, and (3) refueling and maintenance operations in the reactor vessel at 400 to 500°C. As a new concept, the AHTR is early in its development.

CONCLUSION

Although the European and USA interests are focused on different baseline concepts (MSFR and AHTR, respectively), large commonalities in basic R&D areas (liquid salt technology, materials) exist. In Europe, since 2005, R&D on MSR has been focused on fast spectrum concepts (MSFR) which have been recognized as long term alternatives to solid-fuelled fast neutron reactors with attractive features (very negative feedback coefficients, smaller fissile inventory, easy in-service inspection, simplified fuel cycle...). MSFR designs are available for breeding and for minor actinide burning.

Available details of status of these reactor systems are described in subsequent chapters.

Projection of energy needs and the indispensability of Molten salt reactors for future energy security

An interesting “crystal gazing” has been carried out by French to foresee the future global energy needs[1]. This is the summary of that projection.

To meet the future global energy needs, nuclear energy’s contribution is going to be significant. To satisfy these power needs, three typical scenarios, based on three reactor types: Pressurized Water Reactor (PWR), Fast Neutron Reactor (FNR) and Thorium Molten Salt Reactor (TMSR), have been considered. Three main deployment options namely(i) the “PWR only”, (ii) the “PWR + FNR”, and (iii)the “PWR + FNR + TMSR” have been evaluated.

“PWR only” scenario

These reactors operate with thermal neutrons, using light water as moderator and coolant. Two types of PWRs namely the current PWR, and the European Pressurized Reactor (EPR) are considered. The characteristics of these reactors are listed in Table 1.

Table 1 Characteristics of the PWR types used in the “PWR only” scenario

	PWR	EPR
Nominal Power (GWe)	1.0	1.45
Load Factor	0.8	0.8
Launching Date	1970	2005
Lifetime	40 years	50 years
Details of the Fuel (per year):		
Type of Fuel	UOX	MOX-UE with Pumultirecycling
Consumed Fuel	27.2 tons	19.7 tons
Fuel Enrichment in ²³⁵ U	3.5 %	4.5 %
Pu quantity in the Fuel	0 kg	285 kg
Pu produced	270 kg	285 kg
Recovered Uranium after reprocessing	26 tons	18 tons

The “PWR only” worldwide deployment scenario and the corresponding evolution of natural Uranium resources are displayed on Figure 1. In this scenario, the electric power produced in 2030 is twice that of today, EPRs replacing the current PWRs. The power installed continues to grow until 2085 with a capacity of 3700 GWe. At this time, the nuclear power generation stops, as a result of the complete exhaustion of the fissile component of the natural Uranium resources. Moreover, this scenario leads to the build-up of a stockpile of 4000 tons of Plutonium whose management will entail proliferation problems. The main

conclusion of this scenario is that sustainable nuclear energy production is impossible using only PWRs, as was expected.

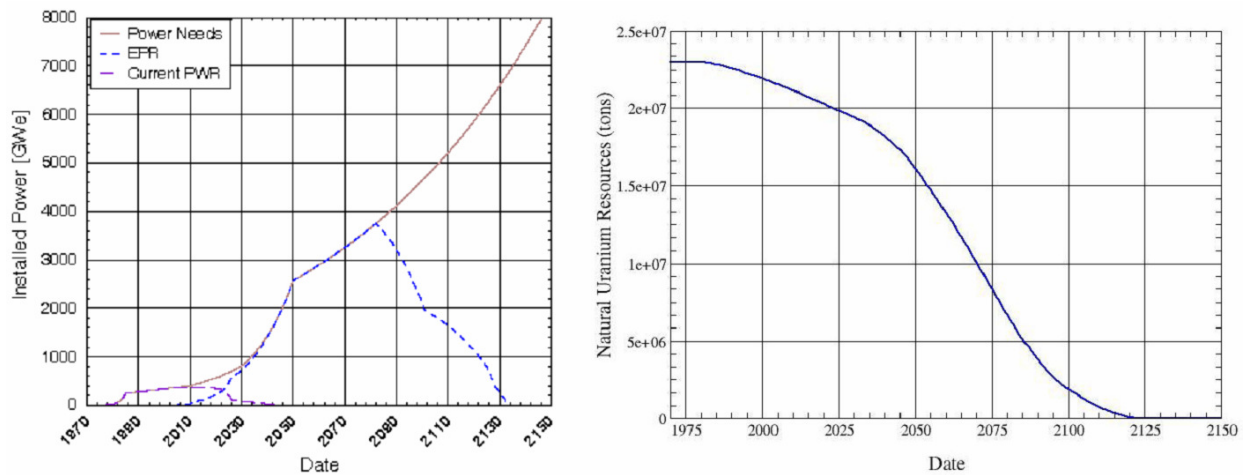


Figure-1 Power available (left) and Evolution of the natural Uranium Resources (right) in the "PWR only" scenario

"PWR + FNR" scenario

The two types of PWR: current PWR and future EPR, are used in this scenario, with a difference: the EPRs operate now with UOX fuel, without multi-recycling the Plutonium which will be needed in the FNRs. These FNRs are breeder reactors are liquid metal cooled and operate on the U/Pu cycle, fuelled with a mix of Plutonium and depleted Uranium. Their estimated characteristics are listed in Table 2.

Table 2 Estimated characteristics of the FNR type used in the "PWR + FNR" scenario

	Liquid metal cooled FNR
Nominal Power (GWe)	1.0
Launching Date	2020
Lifetime	50 years
Fuel Loading/Unloading Frequency	5 years
Fuel Cooling+Reprocessing Time	5 years
Details of the Fuel (per load):	
Depleted Uranium	48 tons
Plutonium	6 tons
Details of the breeding (per year):	
Depleted Uranium Supply	1 ton
Plutonium Bred	300 kg

The worldwide deployment scenario "PWR + FNR" is shown on Figure 2. The corresponding evolutions of natural Uranium resources and of the stock of

Plutonium are displayed on Figure 3. In this scenario, the stock of Plutonium produced in the PWRs is large enough to start the first FNRs. The FNRs will be dominant from 2075 on and their breeding capacity allows then the full nuclear energy deployment, provided EPRs continue to operate.

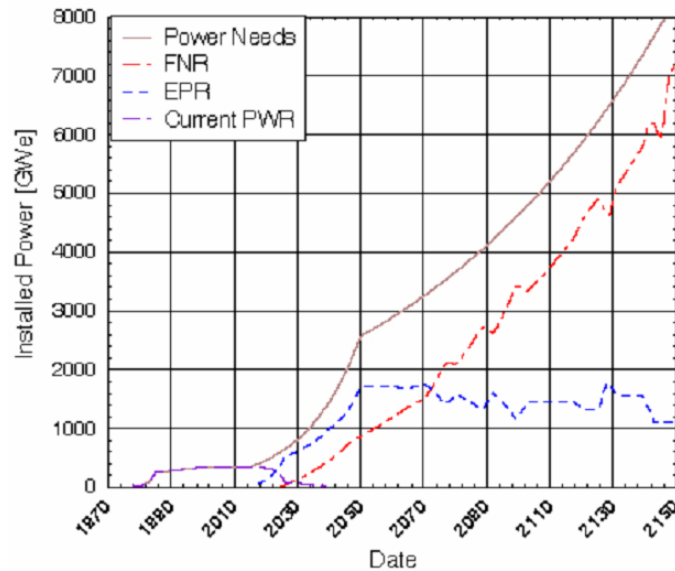


Figure-2 Power available in the "PWR + FNR" deployment scenario

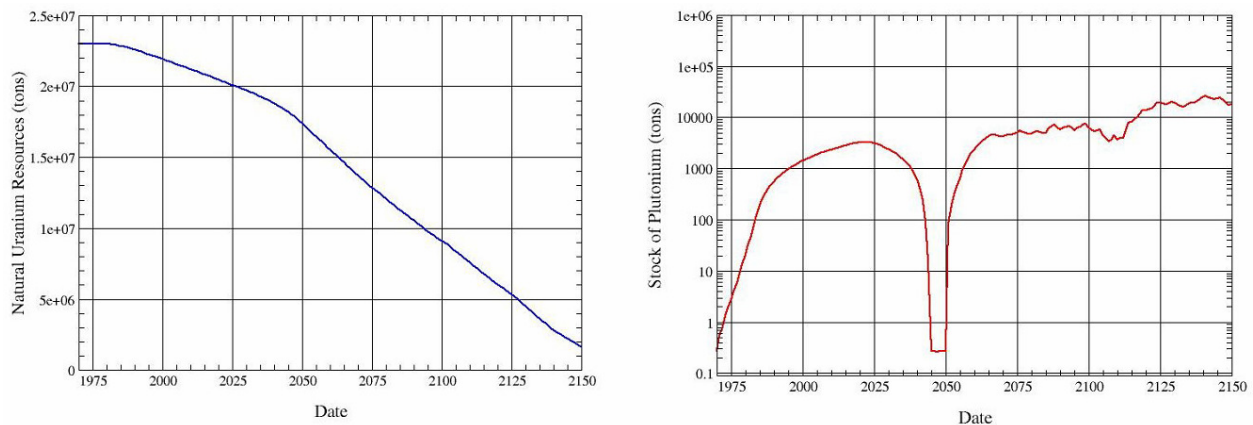


Figure-3 Evolutions of natural Uranium Resources (left) and of the stock of Plutonium (right) in the "PWR + FNR" scenario

A stock of 25000 tons of Plutonium in 2150 is predicted. This large accumulation of Plutonium could cause proliferation difficulties, and this problem cannot be simply resolved by adjusting the FNR breeding rate while satisfying the deployment requirements. Further, the Plutonium produced by the PWRs and available before the launching of the first FNRs is not sufficient for an FNR-only

deployment. The operation of Plutonium-producing EPRs is necessary all through the scenario, leading to the consumption of 80% of the natural Uranium resources in 2150, as shown on Figure 3. As a consequence, this scenario is not suitable for a sustainable deployment of nuclear power, even though the world power needs can be satisfied in the medium term.

“PWR + FNR + TMSR” scenario

The Molten Salt Reactors are based on the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle. As the fissile material ^{233}U required will have to be produced by adding solid Thorium blankets in the EPRs and FNRs. FNRs are now consuming Plutonium and producing ^{233}U . The modified characteristics of these reactors are listed in Table 3. The characteristics of the iso-breeder TMSRs presented in the first section of this paper are summarised in Table 4.

Table 3 Characteristics of the EPR and FNR producing ^{233}U

	EPR with Thorium Blanket	FNR with Thorium Blanket
Type of Fuel	UOX	Depleted Uranium and Plutonium
Fissile material in the fuel	4.9 % (^{235}U)	11 % (Pu)
Details of the inventories (per year):		
Thorium Fuelling	133 kg	500 kg
^{233}U Production	+ 133 kg	+ 500 kg
Plutonium Production	+ 170 kg	- 200 kg

Table 4 Characteristics of the TMSR

	TMSR
Nominal Power (GWe)	1.0
Launching Date	2030
Lifetime	50 years
Details of the Fuel (per unit):	
^{233}U	1.6 tons
Thorium	58 tons
Liquid Thorium Blanket: Thorium Quantity	21 tons

The worldwide deployment scenario “PWR + FNR + TMSR” is shown on Figure 4. The corresponding natural Uranium and Thorium resource evolutions and the stocks of Plutonium and ^{233}U are displayed on Figure 5. The electric power produced in 2030 is twice that of today, EPRs and FNRs replacing the current PWRs. In the meantime, the stock of ^{233}U produced both in EPRs and in FNRs is large enough to start the TMSR systems. These TMSRs will be dominant as early as 2040 and, as for the FNRs previously, their breeding capacity allows successful nuclear

energy deployment. From 2080 on, EPRs are ending their production. Power generation is assumed by FNRs and TMSRs: the transition to the sustainable systems is complete.

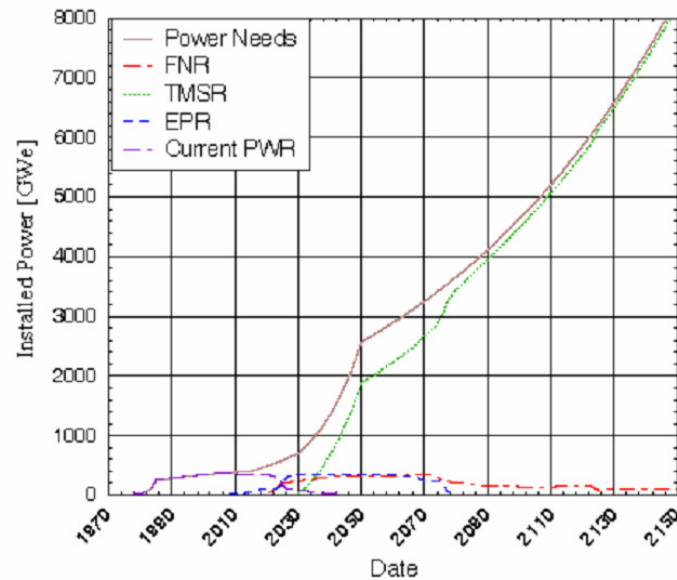


Figure-4 Power available in the "PWR + FNR + TMSR" deployment scenario

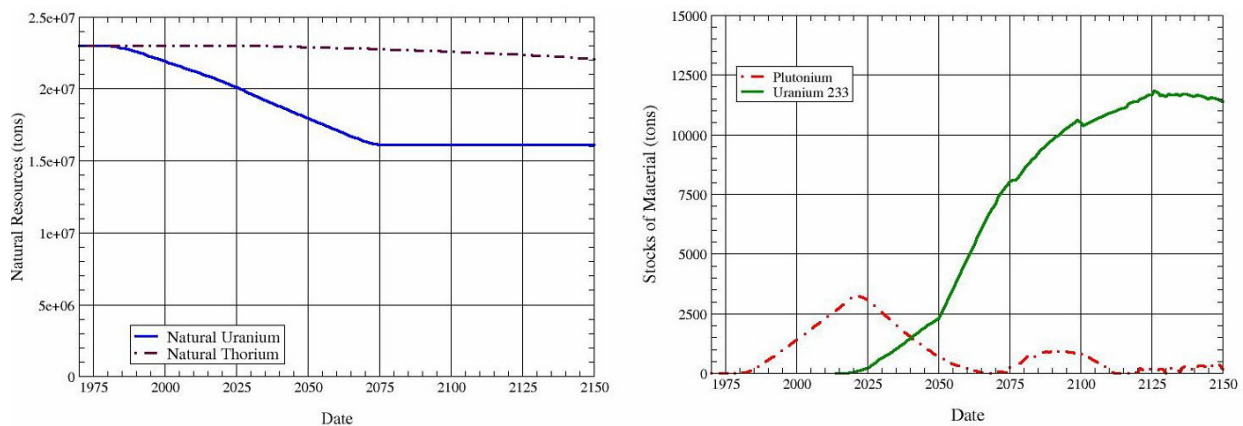


Figure-5 Evolutions of natural Resources (left) and of the stocks of Plutonium and 233U (right) in the "PWR + FNR + TMSR" scenario

In order to consume the stock of Plutonium produced in the PWRs, initially a higher priority for the start-up of an FNR than for a TMSR is envisaged. Figure 5 (right) shows that the stock of Plutonium accumulated before the beginning of FNR launching is consumed in 2150. Figure 4 and Figure 5 (left) show that the worldwide power needs are satisfied all through the scenario without using up all the natural resources: only a third of the natural Uranium and a negligible part of the natural Thorium resources are necessary for the entire timeframe of the

deployment. Assuming a stop of the nuclear energy production, this entire scenario can be restarted at any time, as less than half of the natural Uranium resources are used. Figure 5 (right) shows a large accumulation of ^{233}U during the deployment. This stock can be reduced according to need by modifying the breeding parameters of the FNRs. This scenario, which combines the three types of reactor, is by far the most successful, as it fulfills the requirements for a sustainable deployment of the nuclear power production.

Reference

1. E. Merle-Lucotte, L. Mathieu, D. Heuer, J-M.Loiseaux, A. Billebaud, R. Brissot, S. David, C. Garzenne, O. Laulan, C. Le Brun, D. Lecarpentier, E. Liatard, O. Méplan, F. Michel-Sendis, A. Nuttin and F. Perdu, *Molten Salt Reactors and Possible Scenarios for Future Nuclear Power Deployment*, The Physic of Fuel Cycles and Advanced Nuclear Systems: Global Developments(PHYSOR 2004), Chicago, Illinois, April 25-29, 2004, American Nuclear Society, Lagrange Park, IL. (2004)

TECHNOLOGY CHALLENGES

A series of issues in the 1970s designs of MSR were identified, analyzed, and evaluated in terms of technical solutions available today. Many but not all of these issues were identified in evaluation and closeout reports of the MSR projects in the 1970s.

Power Cycles

When MSRs were being developed in the 1960s, the only demonstrated power cycle for the large-scale conversion of heat to electricity was the steam (Rankine) cycle; thus, early MSRs (and liquid metal fast reactors) were designed with steam power cycles. The coupling of an MSR with a steam cycle resulted in a series of technical challenges. Since that time, gas Brayton power cycles have been developed by the aircraft industry and are now widely used in the utility industry, with natural gas as the preferred fuel.

Direct and indirect Brayton cycles are also being developed for various high-temperature reactors. Indirect multi-reheat nitrogen or helium Brayton, 5 cycles offer major economic and technical advantages relative to steam cycles for electricity production using MSRs.

- **Efficiency.** MSRs are naturally high-temperature reactors. Depending upon the choice of salt, the freezing points are between 320 and 500°C. The heat transfer properties (viscosity, thermal conductivity, etc.) improve rapidly with increasing temperature. Consequently, the detailed 1000-MW(e) conceptual design of the MSR had a reactor-core fuel-coolant exit temperature of 705°C. However, because of corrosion and other constraints in steam cycles, peak steam cycle temperatures are between 500 and 550°C. In the 1960s designs, high temperature heat was inefficiently dumped to lower temperatures to match what the steam cycle could tolerate. This process reduces heat exchanger sizes but has a large penalty in terms of efficiency. In contrast, many Brayton cycles operate above 1000°C. The adoption of closed helium or nitrogen Brayton power cycles enables the power cycle to efficiently use the high temperature heat generated by the MSR. *This capability allows a 15% improvement in electrical power output without changing the temperatures of the fuel salt exiting the reactor core.*

- **Freeze protection.** Salt coolants must be kept sufficiently hot to ensure good heat transport and avoid freezing of the molten fuel salt and the liquid salt in the intermediate heat-transport loop. With a steam cycle and the lower temperatures, special design features must be used so that feed water does not freeze the salt. With the higher-temperature Brayton cycle, freeze protection is greatly simplified.

- *Tritium control.* In an MSR, tritium is generated as a fission product and may be generated by coolant activation. Unlike solid-fuel reactors, the tritium in an MSR is highly mobile in the salt and tends to diffuse through the high temperature heat exchangers into the working fluid of the power cycle. At the time the MSR program was cancelled, tritium control was considered the largest remaining engineering development challenge, because any tritium that entered the steam cycle resulted in tritiated water. Isotopically separating tritiated water from nontritiated water in the steam cycle is difficult and expensive. The MSR program partly developed the use of a fluoroborate coolant salts in the secondary heat transfer system to trap the tritium. While technically workable, such systems are potentially complex and expensive for high levels of tritium trapping. Adoption of a Brayton cycle provides an alternative tritium trapping option where the tritium is removed from the helium in the Brayton power cycle. This is potentially a high performance low-cost option based on demonstrated inexpensive methods to remove tritium gas or tritiated water from helium.

Helium-cooled high-temperature reactors produce tritium from nuclear reactions with ^3He and from leaking fuel; consequently, these reactors are equipped with systems to remove the tritium from the helium.

- *Chemical reactions.* Molten salts do not chemically react with nitrogen or helium. However, these molten salts will slowly react with steam over time. The reaction rate is many orders of magnitude slower than for sodium and water. Changing from a steam cycle to a gas Brayton cycle eliminates this class of challenges. Closed Brayton power cycles using helium are being developed for the modular high-temperature gas-cooled reactors (MHTGRs) for which helium is the coolant gas. A prototype helium-cooled MHTGR is being constructed in South Africa with a helium Brayton power cycle. Additional technology development would be required for an MSR; however, the closed Brayton cycle technology is transitioning to a commercial technology.

Cross-flow plate heat exchangers

These class of cross-flow plate heat exchangers are in one piece in height and vertically divided in 3 to 5 parts of $\ell=10\text{cm}$ each. The total height of these heat exchangers is equal to the height of the fertile blanket. Only 90% in height and in width are assumed to be active to take into account the structures. The thermal exchange capacity depends on the thermal conduction in the thickness " e_s " of the laminar layer of the salt and of the secondary coolant. (See Figure 1)

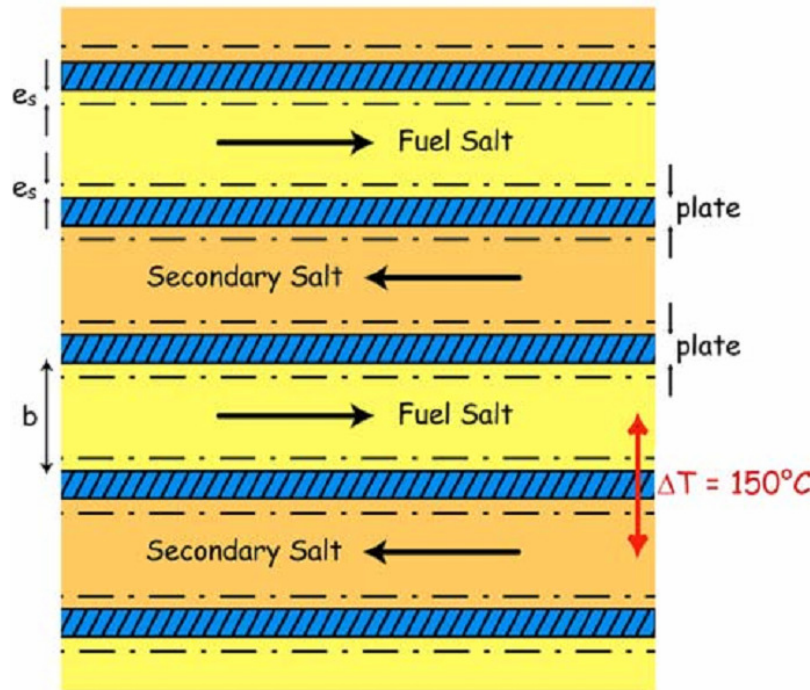


Figure 1 Partial schematic vertical section of the plate heat exchangers, the blue striped areas representing the plates. The temperature differential of 150°C between the primary and the secondary coolants is indicated.

Fuel Inventory

In an MSR, the fuel salt circulates from the reactor core to the intermediate-loop heat exchangers and back to the reactor core. Heat is produced in the core and subsequently dumped in the heat exchangers. Thus, a significant fraction of the fuel salt is outside the reactor core in the heat exchangers. Historically, MSRs have been designed with various tube-and-shell heat exchangers. In the last decade, compact plate-fin and printed circuit high temperature heat exchangers have been developed for the aircraft, chemical, and offshore-oil industries. The adoption of compact heat exchangers drastically reduces the molten fuel salt inventory in the heat exchangers and may reduce the inventory of fuel salt in the reactor by up to 50%. There are major benefits in using such heat exchangers.

- **Fuel salt inventory.** Reducing the fuel inventory reduces both fuel salt costs and nonproliferation risks, because the total fissile inventory in the nuclear system is decreased.
- **Fuel salt processing.** In an MSR, volatile fission products (including xenon) are removed continuously, which creates a large parasitic neutron sink in solid-fuel reactors. For nonvolatile fission products, the fuel salt is processed online or off-line, depending upon design goals. Reducing the salt inventory reduces the quantities of salt to be processed.

- *Heat exchanger size.* The size of the heat exchangers is reduced by a factor of 3 or more. This reduction has major economic implications because the primary heat exchangers have fuel salt flowing through them on one side and clean salt flowing through on the second side. The fuel salt, which contains the fission products and actinides, is highly radioactive. In an MSR, the reactor vessel and primary heat exchangers are located in a hot cell. Reducing the size of the heat exchanger significantly reduces the size of the hot cell, its support equipment, and the reactor building.
- *Tritium control.* The aircraft and other industries have developed compact heat exchangers with buffer gas zones to separate different fluids that may react explosively—such as hot gases vaporizing fuels in aircraft. The same technologies enable trapping of tritium from the primary system in the heat exchanger. While this may not be important for electricity production when using Brayton cycles that allow trapping of tritium, it is another option for tritium trapping if the MSR is used for hydrogen production where high temperature heat is required for the thermochemical hydrogen production cycles.

The advanced heat exchangers are commercial products used in industry. These heat exchangers are being considered for use in high-temperature heliumcooled reactors⁶ and in the transport of heat⁷ from high-temperature gas-cooled reactors to hydrogen production plants using liquid-fluoride-salt heattransport systems. Additional work is required to fully evaluate their use in MSRs.

Noble Metal Plate-Out

In an MSR, fission products are generated in the molten salt. Most of the fission products form stable fluorides that dissolve in the salt. Noble and seminoble metals (e.g., Nb, Mo, Tc, Ru, Rh, Pd, Ag, etc.)³ form multi-atom clusters in the molten salt and ultimately plate out on metal surfaces such as those of heat exchangers or are vented to the off-gas system. The noble metals produce significant decay heat. If the plate-out is excessive, the decay heat from the noble metals may damage the heat exchangers via overheating should a loss of cooling occur. At the end of the MSR projects in the United States in the early 1970s, the assessments indicated that plate-out was not likely to be a major safety issue (potential for noble metals to escape to the environment). However, it had the potential to be a significant design and operational issue. In the last decade there have been major advances in this area.

- *Better understanding of the physical processes:* The R&D challenges are to understand the plateout mechanisms and to test methods for removal of the metal atoms. A major problem has been the difficulty in generating a molten salt with noble metal atoms that can be used to study plate-out and removal

mechanisms. Generating molten salts with noble metal atoms using a molten salt test reactor or an irradiation loop is extremely expensive and involves highly radioactive systems. Recently the French have successfully developed laboratory methods to generate molten salts with noble metal atoms in nonradioactive systems. This development should enable more rapid progress in understanding and development of technologies to remove noble metal atoms from molten salts.

- *New materials.* Plate-out depends upon the surface characteristics. Earlier work showed that noble metals preferentially plated out on metal surfaces relative to carbon surfaces. The potential use of carbon-carbon composite heat exchangers, rather than metal heat exchangers, may significantly reduce noble-metal plate-out on the heat exchangers—the thin-walled reactor component with most of the reactor surface area and most sensitive to decay heat when cooling systems are not operating. Slowing noble-metal plate-out in the primary system provides the time for the salt cleanup systems to remove a larger fraction of the noble metals from the salt.

- *Improved separation methods.* There are several potential methods to remove noble metal from molten salts. The molten salts can be purged with inert gases, with the noble metals preferentially forming aerosols that can be filtered from the gas stream. Newer options include ultra-high-surface-area metal or coated carbon-foam matrixes designed to preferentially encourage plate-out of noble metals in the salt cleanup system.

These developments may allow successful resolution of issues regarding noble metal plate-out issues within several years and help determine (1) whether a significant challenge exists and (2) what the preferred options are for control of noble metal fission products.

Fuel Storage

The use of MSRs will require the storage and transport of MSR fuel salts with and without uranium. The MSR projects in the 1960s did not identify any issues with long-term storage of the fuel salts; however, no long-term tests were conducted. Since that time, events⁸ have revealed challenges in the long-term storage of highly radioactive fuel salts in solid form. The MSRE, an 8-MW(t) test reactor, was shut down and placed in storage in 1969, with the fuel salt (including its uranium) dumped to drain tanks. The fuel salt was stored as a solid at ambient temperatures. In 1994, a gas sample taken from the MSRE off-gas system (which remained connected to the fuel and flush-salt drain tanks) showed the generation of fluorine from the fuel salt and the partial transport of uranium (in the form of UF₆) from the salt into the off-gas system. Molten fuel salts in high radiation fields do not release fluorine, because the fuel salt is an ionic solution with very rapid recombination rates.

However, it is now known⁹ that if the fluoride fuel salt is a *solid at a temperature significantly below its melting point*, radiation can cause the partial decomposition of the salt, release free fluorine, and result in the formation of UF₆. The 30-year storage of MSR fuel salt (an unintended large-scale longterm experiment in fuel-salt storage) and the subsequent remediation program now provide the basis to understand (1) what happens when frozen fuel salts are stored for multi-decade periods of time, (2) the requirements for safe long-term storage, and (3) alternative methods to ensure safe storage.

High-Level Waste (HLW) Form

In an MSR, the fuel is a fluoride salt. Ultimately, the fission products must be removed and disposed of as HLW, while the fissile materials are recycled. This process requires the chemical conversion of the fission products from a fluoride chemical form chosen for in-reactor operations to a repository-acceptable waste form. In the 1960s, no significant work was done to develop such an HLW form. Since the 1960s, however, a variety of other nuclear processing facilities have generated fluoride waste forms. Laboratory studies have been conducted on how to produce high-quality waste forms from many of these fluoride waste streams. Radioactive fluoride waste streams have been generated from (1) processing of spent nuclear fuel (SNF) in the Idaho Chemical Processing Plant, (2) plutonium processing in the weapons complex, (3) development and use of fluoride volatility processing to recover uranium from SNF, and (4) molten salt processing. Two approaches have been partly developed to convert fluoride waste forms to an acceptable form for repository disposal. Either approach potentially provides a basis for development of an MSR waste conversion and solidification process.

- *Conversion to nonfluoride waste forms:* Several processes have been partially developed to convert fluoride waste forms to traditional nonfluoride waste forms. The glass material oxidation and dissolution system (GMODS)^{3, 10} converts fluoride wastes into non-fluoride-containing borosilicate glass—the traditional HLW glass. Other processes produce phosphate waste forms.

- *Fluoride HLW form:* Several potential waste forms contain significant fluorides and may meet the requirements for a repository-acceptable waste form. Borosilicate glasses containing fluorides are a leading candidate for processing the fluoride HLW at the Idaho site¹¹; however, the viability of a fluoride-containing borosilicate glass is strongly dependent upon the chemical composition of the initial HLW form. Other researchers¹² are examining fluorapatites as a waste form. Examples of fluorapatites include Sr₁₀(PO₄)₆F₂ and Sr₈CsNd(PO₄)₆F₂.³

A fully developed process to convert fluoride HLWs into repository-acceptable waste forms does not currently exist. However, several candidate processes and waste forms have been partly developed in the last 30 years.

Peak Reactor Temperature

The peak temperature of an MSR is limited by the materials of construction. The developmental work on MSRs resulted in the development of a modified Hastelloy-N, a high-nickel code-qualified alloy suitable for MSR service that allows peak temperatures to $\sim 750^{\circ}\text{C}$. In the longer term, higher temperatures are highly desirable to (1) improve efficiency in the production of electricity, (2) provide the high-temperature heat required for hydrogen production, and (3) allow the use of higher-meltingpoint fuel salts that may provide major fuel cycle advantages. While there are many hightemperature alloy options for systems with clean fluoride salts, an MSR with dissolved uranium and other species presents special challenges. It is the uranium and certain fission products (not the fluoride salt itself) that primarily determine corrosion rates. In these systems, the corrosion rates are very low with the use of high nickel alloys; however, such alloys lose strength at higher temperatures. Long-term experience shows carbon-based materials to be compatible with molten salts at temperatures of 1000°C . Short-term tests have shown graphite to be compatible with molten salts at temperatures to 1400°C . Carbon-carbon composites are presently being developed for many industrial applications (pumps, heat exchangers, etc.) and have already been developed for use in high-temperature reactors, particularly for in-core high-temperature applications (control rods, core support structures, etc.). Carbon-carbon composites¹³ are potentially an enabling technology for very high temperature MSRs. However, there are major technical uncertainties including joining technologies. If these uncertainties can be overcome, large-scale development work and demonstrations would be required before these materials can be considered for major safety-related components such as reactor vessels. This is a new long-term materials option that did not exist 30 years ago.

CONCLUSIONS

MSRs were developed in the 1950s and 1960s. The large-scale R&D efforts yielded a workable reactor concept but a reactor with significant operational and other challenges. In the last three decades, there have been major advances in technology. A technology gap analysis has identified potential solutions for many of the technological challenges that were identified in the 1970s and that may significantly lower the capital cost of the MSR. The commercial viability of the MSR has improved both in absolute terms and in comparison with other reactor concepts. However, significant

work is required before definitive conclusions can be made about the economics, advantages, and disadvantages of the MSR relative to those of other advanced reactor concepts.

Chemistry aspects of MSRs

Table 1 summarizes some essential characteristics and performances of different MSR concepts.

Table 1 List of MSR concepts, breeder and burner

Family	Concepts	Fast/ Thermal	Fuel Cycle	Thermal power (MW)	Comments
MSR- Breeder	MSBR	T	$^{233}\text{U}/\text{Th}$	2250	Reference breeder concept BR > 1.05 Feedback reactivity coefficient > 0 (slightly)
	AMSTER-B	T	$^{233}\text{U}/\text{Th}$	2250	BR > 0.95
	REBUS	F	U/Pu	3700	
	FUJI	T	$^{233}\text{U}/\text{Th}$	150-200 electrical	
	TMSR	T or F	$^{233}\text{U}/\text{Th}$	2500	BR < 0 Feedback reactivity coefficient < 0 (T and F)
MSR- Burner	AMSTER-I	T	U-Pu-MA	2250	
	SPHINX	F	Pu-MA	1208	
	MOSART	F	Pu-MA	2400	Feedback reactivity coefficient < 0

A screening logic for selecting molten salt fuels and coolants has been propounded by Grimes[1,2]. The “ideal” molten salts and coolants

1. Should have thermal neutron-capture cross-sections of < 1 barn. (not important for secondary coolants)
2. exhibit chemical stability at T > 800°C
3. are stable under intense radiation
4. should melt at useful temperatures (< 525°C) and are not volatile
5. are compatible with high-temperature alloys and graphite
6. dissolve required quantities of fertile and fissile material (for actinide-molten salt)

In addition, the salt must be suitable for fuel reprocessing (full scale or limited). Further, the rationale of selection is also decided by the parameters as indicated below:

Parameter	Rationale
Physical behaviour	Large heat capacity, low melting point, high boiling point, density, viscosity
Chemistry	Chemical stability, Corrosion (clad, piping, reflectors), Chemical reactivity (water, air..), Actinide solubility, Salt clean-up
Core physics	Small capture cross section, non-moderating, low neutron activation, stable under irradiation, reactivity
Safety	Feedback reactivity. Toxicity, optical transparency, in-service inspection
Economics	Cost of salt components, Maintenance, Component cost, Resource availability, Compatibility with structural materials

Useful salt compositions are shown in Table 2 with some factors than can be viewed as stand-alone parameters for screening candidates. Melting temperature is important in all cases. Vapour pressure should be minimized (< 1 mm Hg) to assure salt stability at high temperature. Neutron capture and moderation effect are important for graphite-moderated concepts.

Table 2: Typical salt compositions with some characteristic parameters

Salt	Melting point (°C)	Vapour pressure at 900°C (mm Hg)	Neutron capture relative to graphite	Moderating ratio
LiF-BeF ₂	460	1.2	8	60
NaF-BeF ₂	340	1.4	28	15
LiF-NaF-BeF ₂	315	1.7	20	22
LiF-ZrF ₄	509	77	9	29
NaF-ZrF ₄	500	5	24	10
KF-ZrF ₄	390		67	3
RbF-ZrF ₄	410	1.3	14	13
LiF-NaF-ZrF ₄	436	~5	20	13
LiF-NaF-KF	454	~0.7	90	2
LiF-NaF-RbF	435	~0.8	20	8

Additionally these molten salts should be proliferation resistant.

The relevance of salt selection depends on the application that is considered. Tables 3 and 4 show candidate salts for the different applications.

Table 3: MSR breeder and burner concepts

Family	F/T	Fuel	Fuel cycle	Concepts	Salt type	Selection criteria
MSR-Breeder	T (epithermal)	Liquid fuel	²³³ U/Th	Primary coolant	LiF-BeF ₂ -(HN) ₄	Low melting temperature Low neutron absorption
				Secondary coolant	NaBF ₄ -NaF (MSBR)	Chemical compatibility with water, low cost
MSR-Breeder	F(Non-moderated)	Liquid fuel	²³³ U/Th	Primary coolant	LiF-(HN)F ₄	Low melting temperature Low neutron absorption and moderation
				Secondary coolant		
MSR-Burner	F	Liquid fuel	Pu-MA	Primary coolant	NaF-LiF-BeF ₂ -(HN)F ₃	Actinide (Pu, MA) solubility
				Secondary coolant	NaBF ₄ -NaF	Low cost, tritium trapping
MSR-Burner	F	Liquid fuel	MA	Primary coolant	LiF-NaF-KF-(HN) ₃	MA solubility > 10 mole%
				Secondary coolant	NaBF ₄ -NaF	Low cost, tritium trapping

Table 4: Liquid salt cooled concepts

Family	F/T	Fuel	Fuel cycle	Concepts	Salt type	Selection criteria
AHTR	T	Solid fuel	U/Pu	Primary coolant	BeF ₂ salts NaF salts	Low melting temperature, low cost
VHTR	T	Solid fuel		Intermediate coolant	LiCl-KCl-MgCl ₂ , KF-KBF ₄ , FLiNaK	Heat transfer efficiency, low cost
SFR	F	Solid fuel	U/Pu	Primary coolant	NaF-KF-ZrF ₄	Low melting temperature, low neutron absorption and moderation
				Secondary coolant	Nitrates, chlorides?, hydroxides?	Low melting temperature, chemical compatibility with water

Chemical behaviour

The chemistry of the molten salt is an important parameter for the choice of the salt composition, complementary to neutronic, reactor physics and economic considerations. The most important "chemistry" criteria for any salt are the following:

- Melting temperature:** The liquid range of the salt must fit with the foreseen application and include a sufficiently wide margin toward freezing (solidification). In addition, the vapour pressure of the salt must be low in the relevant temperature range.
- Solubility for actinides (for fuel only):** The fuel salt must be able to contain the required amount of fissile heavy nuclides, again with a sufficient margin, in this case to avoid precipitation of the fissile elements.
- Physico-chemical properties:** Since the prime goal of the salt in any application is the transport of heat, its thermohydraulic behaviour is crucial. The flow of the salt is determined by the viscosity and the density, whereas the heat transfer is determined by the heat capacity and the thermal conductivity.
- Chemical reactivity:** A chemical stability with respect to other fluid media (e.g. fuel salt-coolant mixing, water, sodium) will be required, depending on the environment in which the salt is employed.
- Tritium control:** The necessity of trapping some portion of the tritium in the secondary coolant should influence the selection process.

Table 5 summarises the importance of these criteria for the various applications considered in this report. It is clear that melting temperature and physico-chemical properties are relevant to all of them, whereas solubility and chemical reactivity should be considered for specific applications only.

Table 5 Design significant criteria for various reactor applications

Reactor type	Neutron spectrum	Application	Melting temperature	Actinide solubility	Physico-chemical properties	Chemical reactivity	Tritium trapping
MSR-Breeder	Thermal	Fuel	X	X	X		
		Coolant	X		X	X	X
	Fast	Fuel	X	X	X		
		Coolant	X		X	X	X
MSR-Burner	Fast	Fuel	X	X	X		
		Coolant	X		X	X	X
AHTR	Thermal	Coolant	X		X		X
VHTR	Thermal	Heat transfer	X		X		X
LFSR	Fast	Coolant	X		X	X (with water)	
SFR	Fast	Heat transfer	X		X	X (with sodium)	

The binary phase diagrams are assessed based on the experimental data, mostly on the solidus, liquidus and phase transition data points. The excess energy of the solutions (in most of the systems only the liquid solution) is optimized in order to obtain the best possible agreement between phase diagrams and measurements. It is of great importance to model the higher order systems. These are extrapolated on a basis of the binary data according to the Kohler symmetric or Toop asymmetric formalism. Thus obtained higher order phase diagrams (ternary, pseudo-ternary etc.) are compared to the experimental data (if these are known) and if necessary adjusted by introducing the ternary excess energy parameters. Considering these criteria and other thermodynamic approaches to eutectic and melting points of the mixtures, and solubility of fuel materials in the salt mixture wherever needed and also taking into account the neutronic considerations for reactor fuels, the following chemical families can be discriminated:

- a) Fluorides of metals with low neutron capture cross sections that can be used in reactor cores, where a moderation effect is not wanted for fast neutron cores. This principally includes most of the alkali metals (^7Li , Na and Rb) as well some alkaline-earth metals (Be, Ca). The system $^7\text{LiF-BeF}_2$ is the reference for this family, eventually extended to $^7\text{LiF-NaF-BeF}_2$. Although zirconium also has a low neutron capture cross section in the thermal part of neutron spectrum, its volatility is relatively high and it can only be considered as salt component in exceptional cases. In non-moderated cores, also KF can be considered as major constituent in the fuel solvent system.
- b) Fluorides for application outside reactor cores, such as LiF-NaF-KF (FLiNaK), NaF-NaBF₄ and KF-KBF₄, for which no neutronic considerations apply.

- c) Chlorides of metals with low neutron capture cross sections that can be used in fast reactor cores. For this application, LiCl, NaCl and MgCl₂ and their mixtures can be considered.
- d) Nitrates can be an alternative to chlorides and fluorides for heat transfer applications at low temperatures. The system NaNO₃–KNO₃ is the probable choice.

Material compatibility

A critical technical distinction between the MSR and other liquid salt applications should be noted. The corrosion rates of systems containing clean liquid fluoride salts with the proper materials of construction are very low; it is the impurities that are primarily responsible for corrosion. Appropriate alloys of construction have been found for MSRs. However, the peak temperatures may be limited to less than 750°C because the same alloys have low strength at higher temperatures. This constraint does not exist for clean liquid salt systems.

The success of a Molten Salt Reactor (MSR) is strongly dependent on the readiness of structural material with long service lifetime at the intended high temperature of use. This includes creep resistance and compatibility with the molten media used as the primary and possibly as secondary coolant. Corrosion is the most unique and critical materials requirement, since many alloying elements commonly used in high-temperature structural alloys exhibit some degree of solubility in molten fluoride salts. The reliance on a protective oxide layer is not practical because oxides are chemically unstable in molten fluoride salt environments. Resistance against high-temperature air oxidation is a further criterion on materials, since the outer surface of the molten salt containment will likely interface with air.

USA (especially ORNL), Europe and former Soviet Union have accumulated significant knowledge in the operation of molten fluoride salt test loops using many coolants (NaF–ZrF₄, LiF–NaF–KF, LiF–BeF₂ etc.) Implementation of two major experimental programs in USA, that used UF₄–ZrF₄–NaF fuel and moderated by BeO, and the Molten Salt Reactor Experiment (MSRE), fuelled with ⁷LiF, UF₄, ThF₄ and BeF₂, also provides valuable information.

Early studies at ORNL on candidate metallic materials showed Ni-based alloys and austenitic stainless steels to be generally promising candidates for molten salt environment. These tests were performed in a temperature gradient system with various fluoride media, at different temperatures (maximum temperature and temperature gradient) and different time durations. Chromium, which is added to most alloys for high-temperature oxidation resistance, is quite soluble in molten fluoride salts. Metallurgical examination of the surveillance specimens showed the corrosion to be associated to outward diffusion of Cr through the alloy. It was concluded that the chromium content shall be maintained as low as reasonably possible to keep appropriate air oxidation properties.

Corrosion in the reactor circuit was controlled by reducing approximately 1% of the UF₄ solute to UF₃, so that the oxidative equilibrium between the chromium of the Hastelloy N container was shifted to the left:



Further postoperative examination showed that fission-product tellurium diffused into the Hastelloy N grain boundaries and caused an unacceptable stress-cracking embrittlement of the metal. Radiation hardening of the Hastelloy N and diffusion of tritium (which was produced by the action of the neutrons on the lithium component of the solvent) were also of concern for future reactor applications.

Corrosion rate is marked by initial rapid attack associated with dissolution of Cr and is largely driven by the impurities in the salt. This is followed by a period of slower linear corrosion rate behaviour, which is controlled by a mass transfer mechanism dictated by thermal gradients and flow conditions. Minor impurities in the salt can enhance corrosion by several orders of magnitude and must be kept to a minimum. Dissolution can be mitigated by a chemical control of the redox in salts for example by small additions of elements such as Be. Corrosion increased dramatically as the temperature was increased and is coupled to plate-out in the relatively cooler regions of the system, particularly in situations where high flow is involved. Tests performed at 815°C especially showed Ni based alloys to be superior to Fe-based alloys. This led to the development of a tailored Ni-based alloy, called INOR-8 or Hastelloy-N, with a composition of Ni-16%Mo-7%Cr-5%Fe-0.05%C, which has successfully functioned in molten fluoride salt at maximum 650°C for 26000 h.

However, during MSRE operation, Hastelloy-N appeared to be sensitive to in-service embrittlement due to two processes. On the one hand, the interaction with the fission product tellurium causes an intergranular attack and, on the other hand, irradiation by delayed neutrons produces helium. Additions of small amounts of Nb and Ti were shown to improve the strength of Hastelloy. Furthermore, it was seen that the control of the molten salt oxidation potential had dramatic effects on the extent of cracking as shown in Figure 1.

EXTENT OF TELLURIUM EMBRITTLEMENT OF HASTELLOY N
IS STRONGLY AFFECTED BY OXIDATION POTENTIAL OF SALT.

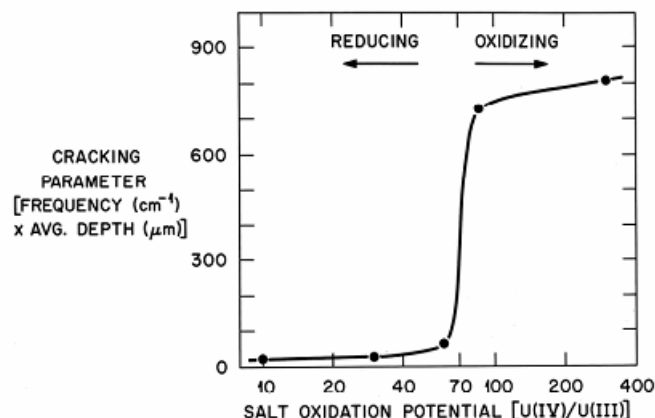


Figure 1 Effect of redox potential on tellurium cracking of Hastelloy N

Russian studies have shown further possibilities for compositional modifications of Hastelloy-N for improving its resistance to embrittlement in high-temperature molten fluoride salt environment. About 70 Ni-base different casts were tested with alloying elements such as W, Nb, V, Al and Cu. Some experiments of corrosion tests of Ni-base alloys in molten Li,Na,Be/F salts with PuF₃ and Te additions were also carried out in Russia. It was concluded that addition of alloying elements can provide resistance against tellurium embrittlement together with keeping the good corrosion properties of the alloy.

The composition of candidate metallic structural materials with Ni-base is in Table 6. Only few of them are alloys with ASME code certification for high temperature application.

Table 6 Alloy composition (in wt%) of MSR candidate structure materials

Alloy	Cr	Mo	W	Ta	Al	Ti	Fe	C	Co	Ni	Nb	Zr
Hastelloy-X	20.5	8	0.2				17	0.05	0.5	Bal.		
Hastelloy-N	6.31	16.1	0.06		0.01	4.03	0.03	0.03	0.15	72.2		
M22	5.7	2	11	3	6.3			0.13		Bal.		0.6
HP-Modified	24						Bal.	0.37		34	1.2	
Nickel-200										99		
Incoloy-800H	19				0.15	0.15	Bal.	0.05		30		
Haynes-230	22	2	14				3		5	57		
Nb-1Zr											98.9	1.1
Haynes-214	16				4.5		3	0.05		75		0.1
Inconel-617	24	10			0.8	0.6	3	0.15	15			
Haynes-242	8	25			0.5		2	0.03	2.5	65		
Hastelloy B-3	1-3	27-32	3		0.5	0.2	1-3	0.01	3	Bal.	0.2	

Corrosion proved to be highly sensitive to impurity contents in the salts, to temperature and to the alloy chemical composition. Chemical composition of the salt, temperature gradient, hydrodynamics are also likely to play a role. Corrosion must be mitigated by using the appropriate combination of materials and salt

chemistry. This implies to select corrosion-resistant high temperature alloys and to chemically control the redox in salts.

From the material point of view, the need is for further assessment of existing materials and the selection of new materials for which less data exist. Nickel being the most resistant element to dissolution in salts, Ni-based alloys such as Hastelloy, are the most promising candidates for application up to 750°C.

Selection of appropriate alloying elements should provide for the resistance to embrittlement by Te-attack and He production. These optimized Ni-base alloys shall be resistant to irradiation damages and be licensed for nuclear power plant applications in accordance with international standards and codes (for example ASME code). Candidate materials with ASME code certification are given in Table 7. From the standpoint of creep strength, there are a number of commercial Ni-based alloys that could be used between 750 and 850°C. However, extensive long-term corrosion evaluation of these alloys in molten fluoride salts is required and the possibility to use coatings may be investigated. Temperature greater than 850°C would require the use of new solutions such as refractory alloys or graphite.

Table 7 Candidate materials with ASME code certification for high temperature applications

	Candidate materials	Metallurgical stability	Irradiation resistance	Fabricability	Alloy maturity	Codified
Coated	9Cr-1MoV	Fair	Good	Good	High	Sect III, VIII
	304	Good	Good	Good	High	Sect III, VIII
	316	Good	Good	Good	High	Sect III, VIII
	347	Good	Good	Good	High	Sect III, VIII
	Alloy 800H or HT	Good	Good	Good	High	Sect I,III, VIII
Monolithic	Hastelloy N	Good	Good	Good	High	Sect III, VIII
	Haynes 242	Good	Adequate	Good	Low	Sect VIII
	Alloy 800H or HT	Good	Good	Good	High	Sect III, VIII

First priority R&D needs in corrosion for metallic materials of primary and secondary circuit are to focus on the pre-selected salts for the different concepts. For fuel salts, interest is on ${}^7\text{LiF}-(\text{NaF})-\text{ThF}_4$ and ${}^7\text{LiF}-(\text{NaF})-\text{BeF}_2-\text{ThF}_4$. For coolant salts, ${}^7\text{LiF}-\text{BeF}_2$ is the main candidate for in-core use and $\text{NaF}-\text{NaBF}_4$ as well as ternary $\text{LiF}-\text{NaF}-\text{BeF}_2$ mixture with melting temperature 315°C for secondary coolant for systems producing tritium. Finally, $\text{NaNO}_3-\text{KNO}_3(-\text{NaNO}_2)$ is cited for heat transfer applications. For these systems, redox control must be experimentally demonstrated and the operating temperature range for acceptable resistance to dissolution has to be established. Molybdenum, which also has a good compatibility with fluorides, was therefore added in solid solution to nickel to provide high temperature creep resistance and hardening. The composition of Cr was tailored on the one side to maintain a good corrosion resistance in gas atmosphere containing oxygen due to the formation of a protective oxide. On the other side, the content of Cr was limited in order to suppress voids formation due to the Cr depletion by dissolution in the molten salt.

Helium produced by neutron capture on the isotope ^{58}Ni dominates the issue of the resistance of the material under irradiation. A modified version of Hastelloy N was designed with an improved irradiation resistance due to a fine dispersion of titanium and niobium carbides. These carbides provide coherent interfaces to the nickel matrix which very efficiently traps He atoms.

One can say that within the temperature range envisioned for molten salt reactors at that time (maximum temperature of 700-720°C), there is a first generation structural material that satisfies requested criteria. However, it was also demonstrated that the maximum temperature allowable for this material is of the order to 750°C. Indeed, beyond this threshold, titanium and niobium carbides are dissolved in the nickel matrix. Due to its evolving microstructure, it would therefore be impossible to preserve the material properties required to address the specificity of molten salt reactors at higher temperature.

Replacing molybdenum by tungsten in such alloys could prove beneficial to reach higher in-service temperature from several standpoints. First of all, tungsten diffusion is roughly ten times slower in nickel than molybdenum diffusion [7]. Therefore, there is correspondingly a better creep resistance expected with a Ni-W solid solution than with a Ni-Mo solid solution. This would help to reach higher in-service temperature. Second, a comparison of the ternary phase diagram of Ni-Mo-Cr with Ni-W-Cr shows that there is only one intermetallic phase with a high Cr content. Close to the solubility limit in the low chromium range, there is no embrittling intermetallic in the Ni-W-Cr system. Instead, there is a phase separation between the solid solution and a pure W α -phase. Table 8 lists some of the suitable candidate materials for various components of MSRs.

Table 8 Candidate structural materials for high temperature applications

	Candidate materials	Salt corrosion resistance	Air corrosion resistance	Long term strength at 1000°C	Highest usage temperature (°C)	Potential MSR, AHTR component usage
Monolithic	Hastelloy N	Excellent	Good	Very good	730	
	Haynes 242	Very good	Good	Very good	540	
	Alloy 800 H or HT	Poor-fair	Good	Very good	980	
Coated	Inconel 617	Needs evaluation	Good	Very good	1000	PM,P,V,HX
	VDM 602 CA	Needs evaluation	Good	Good	1000	P,V,HX
	Alloy 800H	Needs evaluation	Good	Good	1000	P,HX
	Haynes 230	Needs evaluation	Poor	Good	900	P,HX
	Hastelloy X or XR	Needs evaluation	Marginal	Good	900	P,HX
	HP modified	Needs evaluation	Good	Excellent	1100	V
Monolithic	Haynes 214	Very good	Good	Good	1000	V,HX,CHX
	MA 956	Very good	Good	Good	?	HX,CHX
	MA 754	Very good	Good	Good	?	HX,CHX
	Cast Ni superalloys	Very good	Good	Good	?	PM

PM= Pump, P= Piping, V= Valves, HX= heat exchanger, CHX= compact HX

Graphite

MSRs may use graphite as a moderator and reflector. The MSRE program used graphite in the core as a neutron moderator, with no observable deleterious reactions after long-term direct contact with molten salt.

There are two technical challenges: the need for stability of the material against radiation-induced distortion and low permeability to salt and gas ingress. Radiation damage that changes the geometry necessitates replacement of the graphite moderator — an expensive maintenance task. For conventional graphite irradiated up to fluences $> 2 \times 10^{22}$ neutrons/cm² ($E > 50$ keV), volume changes lead to formation of cracks sufficiently large for salt intrusion. To address this, fine grained nuclear grade graphite must be used for in-core applications and cross-cutting can be found with the development of graphite for gas cooled reactors.

In MSR, cracks and crevices in the graphite can allow the xenon gas to remain in the reactor core where it is a strong neutron absorber and where it will significantly reduce the breeding ratio of the reactor. Low porosity levels are thus desirable in order to prevent salt ingress. The approach taken in the MSBR program was to seal the graphite surface with pyrocarbon to exclude xenon. No work has been done in several decades to lower the permeability of graphite for MSR service.

If these technologies do not meet future MSR requirements, new methods for sealing graphite need to be developed. Appropriate tests under irradiation must be conducted to demonstrate the sealing of graphite surfaces. The R&D needs for graphite as moderator and/or reflector are thus mainly directed toward qualification of radiation-stable grades, sealing technology to reduce gas ingress and fabrication issues.

Materials for fuel processing units

Preliminary considerations on problems and required tests for materials of fuel processing components and systems are briefly listed in Table 9.

Table 9 Fuel processing materials development

Concept	Process	Material	Problems and required tests
MSRE	U-fluorination (UF ₆ , HF)	1) Ni-base alloys 2) Mo, graphite, Ta, braze alloys	1) Frozen salt layer 2) Chemical compatibility tests, capsule & thermal convection, fabrication joining techniques
MSBR	Reductive extraction		
TMSR	MS / Bi-Li-Th		
MOSART	MS / Bi-Li		

Materials for large scale components

Material-molten salt compatibility and structure integrity depend as a rule on the component scale. In the framework of the MSRE, primary circuit components (reactor structure, salt pump, pipelines, salt-air heat exchanger etc.) and consequently their material compatibility with the fuel salt were tested in the

scale corresponding to the thermal power rate of 8 MW. No tests were performed on salt-water/steam and salt gas (CO₂, N₂ etc.) heat exchangers neither at small nor at large scale. Salt composition and temperature distribution also influence the neutronics aspects of MSRs. This would in turn has a direct bearing on the safe operation of the reactor. These are discussed in physics section.

Neutronic aspects

Preliminary physics considerations

The different kinds of reactors using molten salt as a fuel or as a simple coolant use a very wide range of neutron spectra, from very moderated to fast spectrum.

Breeding in thermal spectrum (MSBR/epithermal TMSR / low power epithermal TMSR) is possible with ²³³U/Th. One advantage of thermal breeders is low fissile inventory compared to fast spectrum (good for safety and deployment speed). To allow breeding, the η value has to be sufficiently higher than 2 (which is the case for ²³⁹Pu and ²⁴¹Pu for fast neutrons but not for thermal neutrons). In the case of ²³³U, breeding is possible in both thermal and fast spectrum as η is more than 2 in both the cases.

Thermal breeders require graphite to moderate neutrons (very low capture, good slowing down cross section) as carbon has a good compatibility with molten salts. The associated problem as mentioned previously, is its weak resistance under irradiation (especially under fast neutron flux) requiring to change graphite every 4 years in the MSBR or 2 years in the epithermal TMSR. This motivates the study of very low power reactors with the objective of 40 years graphite life with the concept called very low power epithermal TMSR.

Efficient breeding needs reasonably fast on-line reprocessing to remove the neutron poison fission products and plough back the molten salt to reactor. Figure 2 shows relative amounts of neutron poison fission products formed.

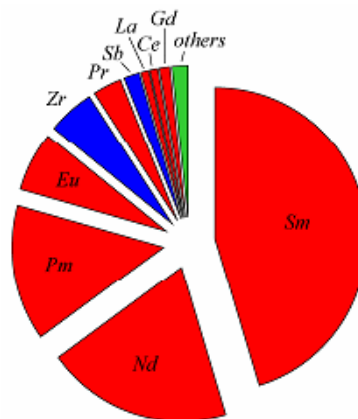


Figure 2 Distribution of the capture rate in the fission products in the core after helium bubbling only for a thermal breeder

Samarium is the most disturbing poison. The following ones (like Neodymium or Promethium) belong to the same chemical family, the lanthanides (red part of the Figures). Among other contributing fission products, Zirconium represents more than 50% of the captures produced by elements other than lanthanides. The Molten Salt Reactor allows the on-line reprocessing of the fuel and is particularly well fitted to thorium breeding which has a tight neutron economy. Fluoride salts are preferred for primary coolants as fluorine has the least neutron absorption cross section among F, Cl, Br. Chlorine can be considered for fast spectrum because capture cross sections are small.

Fission products have very low capture in fast spectrum. Therefore, a lower reprocessing rate is needed for fast neutron concepts.

Breeding in fast spectrum

For breeding in ^{238}U -Pu cycle (REBUS, LSFR), a fast spectrum is necessary because the neutron excess value per Pu fission rapidly increases with the neutron energy. If the spectrum is not hard enough, breeding is no more possible. Fluorine (F) has a higher scattering cross-section than chlorine (Cl) in the fast range (10^4 to 10^6eV) with high scattering resonance. Furthermore, F is twice lighter than Cl so that its slowing down power per collision is higher. That is why chlorine (Cl) is better than fluorine (F) as far as breeding in ^{238}U -Pu cycle is concerned.

For breeding in ^{233}U -Th cycle (non-moderator TMSR), an intermediate/fast spectrum is enough for breeding in Th/U cycle because ^{233}U neutron excess per fission is rather independent from the spectrum compared to Pu. Thermalisation by fluorine (F) does not impair the breeding of ^{233}U , therefore a fluoride salt can be used for this application. Choice of secondary salt constituents for Th- ^{233}U breeding, where spectrum hardness is not required, any addition of Li, Be, Na, Mg, K, Ca, Ti, Rb, Sr, Zr can be considered.

The neutronic irradiation damages to the structural materials modify their physicochemical properties through three effects: the displacements per atom, the production of Helium gas via nuclear reactions in the materials, and finally the transmutation of the Tungsten (component of the Ni-based alloy) to Osmium through nuclear reactions. These damages have been computed for the axial reflectors in the core for the medium case corresponding to a fuel salt volume of 18 m^3 .

The irradiation damages are inversely proportional to the fuel salt volume of the reactor, thus logically favoring the larger MSFR configurations. These damages are dominated by Helium production through the (n, α) reaction on ^{58}Ni . Two solutions may be considered: either regularly replacing the superficial layers (around 15 cm) of the axial reflectors and the fertile blanket container; or selecting another material without boron or ^{58}Ni . The latter solution may prove more expensive but will be reserved to the more exposed areas. The effects of Helium on the structural materials (maximal acceptable amount, diffusion) have to be thoroughly studied

by specialists to confirm our preliminary conclusions. Likewise, the effects of the transmutation to Osmium of the Tungsten contained in the structural materials have to be studied for a better understanding of the long-term materials resistance.

Optimization of the burning capacities of the system favours the configurations with the smaller fuel salt volumes. As a conclusion, the MSFR configurations containing medium fuel salt volumes, between 15 and 20 m³, are a good compromise to satisfy all viewpoints (deployment, safety, materials damages and burning capacities).

Nonmoderated burner

Non moderated MSR is efficient for actinide burning because fast spectrum is favourable for TRU burning in general. This is due to the probability of fission per neutron absorbed which is much higher in fast spectrum than in thermal spectrum, especially for isotopes non fissionable by thermal neutrons (Figure 3).

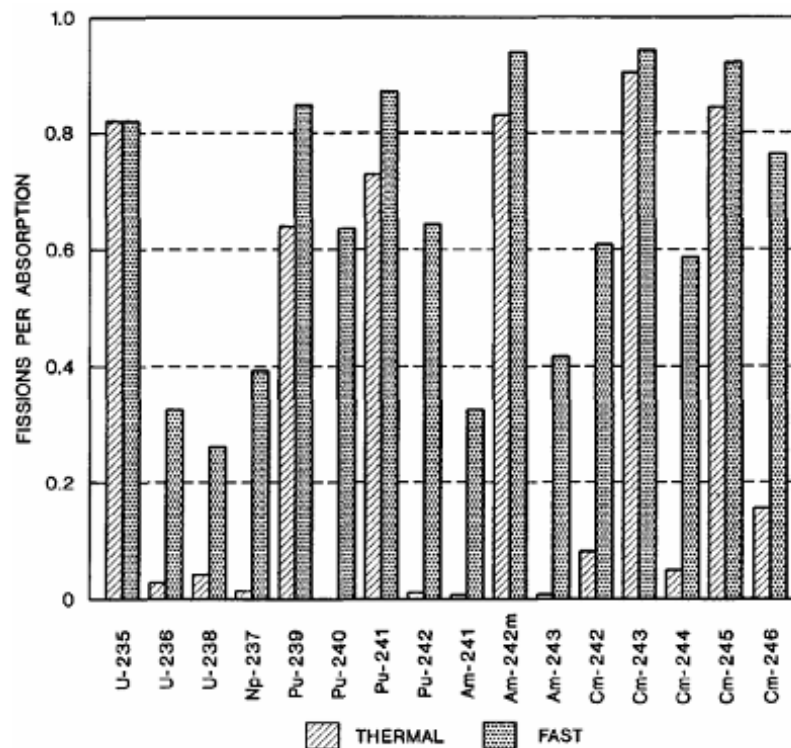


Figure 3 Probability of fission per neutron absorbed of some heavy nuclei (Thermal/Fast)

The main constraint for this type of reactor is to remain below the solubility limit of TRU in the salt.

Main conclusions on the different concepts

The main neutronic characteristics for the different concepts are listed in Table 10. One can see that the different goals pursued by the different types of reactors (burning, breeding) and different choices (liquid/solid fuel) lead to very distinctive choices for salt composition. The most attractive concepts are two non-moderated concepts, one breeder (non moderated TMSR) and one burner (MOSART).

Table 10 Neutronic characteristics of the reactor

Reactor type		MSR						Solid fuel, liquid salt cooled	
Main function		Breeder				Pu+MA burner		Breeder	High t
Fuel cycle		Thorium			Uranium	Inert support	Thorium support	Uranium	Uranium
Spectrum		Epithermal		Fast	Fast	Fast	Fast	Fast	Thermal
Name		TMSR epithermal & low power	TMSR epithermal moderated	TMSR non moderated	REBUS-3700 non moderated	MOSART non moderated	TMSR Pu+MA started	LFSR	LSPBR
Unit power (MWe)		150	1000	1000	1540	1000	1000	1000	1000
Reference salt		78LiF-22HNF ₄	78LiF-22HNF ₄	82.5LiF-17.5HNF ₄	38UCl ₃ -7TRUCl ₃ -55NaCl	58NaF-15LiF-27BeF ₂	82.5LiF-17.5HNF ₄	NaF-KF-ZrF ₄	66 LiF-34BeF ₂
Fuel cycle	Burnup at eq.		0.7 at%	2.4 at%	4.8 at%	11.6 at%	2.4 at%	10 at%	10 at%?
	Reprocessing time (efpd)	500	150	418	1500	300	418	~1500	No reproc.
	HN reprocessed kg/d at eq.	194	334	100	47	21	100		X
	Fertile feed kg/TWhe	100 kg Th	100 kg Th	100 kg Th	100 kg U _{dep}	0	100 kg Th	100 kg U _{dep}	
	Fissile feed kg/TWhe	0	0	0	0	100 kg Pu+MA	0	0	
	Initial HN mass kg/GWe	625,000	50,160	41,800	74,600	2,746	41,800		
	Initial TRU mass kg/GWe	0	0	0	11,700	2,746	41,800		
	Initial 233U mass kg/GWe	14,525	1,920	4,650	0	0	0		
	Equilibrium HN mass kg/GWe	625,000	50,160	41,800	74,600	2,746	41,800		
	Equilibrium TRU mass kg/GWe	Low	18.5	650	11,700	6,280	650		
	Equilibrium 233U mass kg/GWe	9,960	1,920	7,500	0	0	7,500		
Graphite	Lifetime years	40	1.75	ZrC reflector	Metal reflector	4	ZrC reflector		
	Wastes kg/d	7	49.3	Low	X	13.7	X		NC
Safety parameters at eq.	Total pcm/K	-0.5	-2.37	-7	-6	-3.7	-5		-8.5
	Coolant pcm/K	-2.0	-2.89	-7	-8	-3.7	-5		-1
	Fuel pcm/K								-7.5
	Moderator pcm/K	1.4	+0.52	X	X	X	X		
	Reflector pcm/K	NC	NC	NC	NC	NC	NC		
Eff. Pcm with moving fuel correction		190	190	190	250	200	190		
Comments								Coolant void effect: +8500 pcm	12 gU/pebble 10% enr.

NC = Not computed

From the Table 10 it may be seen that in non-moderated Thorium breeder MSR (non-moderated TMSR) the reference salt is 82.5%LiF-17.5% HfF_4 in molar proportion. There is no neutronic penalty to add a third component to the salt e.g. NaF, BeF_2 or CaF_2 (or even small quantities of Mg, K, Ti, Rb, Sr, Zr) to improve the chemistry (for reprocessing and compatibility with structural materials). The safety coefficients are excellent (salt temperature coefficient -7 pcm/°C). The initial fissile inventory is higher than for thermal breeder (4.7 t ^{233}U /GWe). The possibility to start these reactors directly with Pu has to be analysed from the chemistry point of view because it has been shown possible with respect to neutronics. The very good potential of this option makes it the reference breeder solution to investigate in future projects

For MOSART, the fuel salt mixtures identified are ternary Li,Na,Be/F and binary Li,Be/F solvent systems with decreased content of beryllium difluoride (25-27 mol%), having satisfactory melting temperatures (T_{melt} (Li,Na,Be/F) $\approx$ 480°C and T_{melt} (Li,Be/F) $\approx$ 530°C), required solubility of actinide trifluorides (> 2 mol%), adequate thermo-physical properties, as well as suitable costs. Considered compositions have very high radiation resistance and relatively low corrosivity to graphite and Ni-Mo alloys. Actinides and soluble fission products can be easily removed from molten LiF- BeF_2 -based mixtures. Optimum spectrum for MOSART is intermediate spectrum of homogeneous MSR without graphite moderator. Due to intensive production of ^{245}Cm in spectrum typical of such a core, it can operate without additional neutron sources. The equilibrium inventory of TRU is 6.3 t/GWe, which remains under the solubility limit. The safety is excellent, with a very negative salt temperature coefficient (-4 pcm/°C). The reactor is fed with TRU coming from PWR spent fuel.

The very good potential of this option makes it the reference burner solution to investigate in the future. Particularly, an interesting point would be to study its ability to burn TRU (MA and excess Pu generated by breeding) coming from solid fuel Fast Breeder Reactors or even only minor actinides from solid fuel reactors. Obviously, the latest scenario will require new fuel solvent systems with higher solubility for MA (e.g. LiF-NaF-KF) compared to Li,Na,Be/F or Li,Be/F MOSART fuel salt matrix.

Reprocessing and fuel salt clean-up

Fuel salt clean-up is a part of MSR fuel cycle technologies. The technological processes used for fuel salt clean-up are generally based on the pyrochemical separation methods and techniques using molten salts. The main reason for the fuel salt clean-up is to provide a composition of the fuel salt in primary circuit within the required limits, which enables to keep the reactor in operation for a long run. The task of the fuel reprocessing is then removal of fission products, which are appreciable neutron poisons, extraction of newly constituted fissile material

or its precursor and finally refueling of the reactor by some fresh fissile and/or fertile material.

The use of individual separation processes could be dependent on the type of fuel composition in MSRs, which could consist of uranium and thorium fluorides, fluorides of transuranium elements or their mixtures. As MSRs can be used as actinide burners or thorium breeders, each of this type has some individual requirements for its spent fuel reprocessing.

The processing of fresh transuranium fuel for MSR working as actinide burner covers the classical reprocessing technologies either based on hydrometallurgical PUREX process and subsequent advanced hydrometallurgical separation methods with final conversion into fluorides, or some advanced pyrochemical technologies like fluoride volatility process. While the solid fuel technology is focused to the separation of individual actinides with a maximum purity, the MSR fuel salt clean-up is focused to the removal of fission products (with exception of U-233) and the individual element separation is not necessary. Also, the choice of carrier molten salt in the solid fuel technology is generally free but the choice of carrier salt in MSR technology is significantly determined by the connection to the MSR physics.

There were at minimum two important problems from the fuel cycle area which were solved during the MSRE program. The first was the cleaning of xenon, krypton and noble metals from the circulating fuel, the other was the substitution of uranium-235 by uranium-233 in the fuel (primary) circuit.

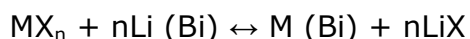
Among the fission products, noble gases (mainly xenon-135) were some of the most neutron poisoning elements in the circulating fuel salt. Therefore the removal of noble gases from the circulating fuel had to be provided when the reactor was in operation. Noble gases have very low solubility in the fuel salt ($\text{LiF}-\text{BeF}_2-\text{UF}_4$), which makes possible to strip them from the salt and reduce the poisoning effect of Xe-135. The removal was provided by sparging the salt with helium, which was then passed through charcoal traps in an off-gas clean-up system. The contact for noble gases (xenon and krypton) removal was produced by spraying of salt through the cover gas (helium) and into the pool of salt in the fuel pump bowl. Argon was also tried for cover gas. When the reactor was operated at the power with argon cover gas, the xenon poisoning at high void fractions was about the same as it was with helium, but the xenon poisoning was higher with argon when only small amounts of gas circulated with the salt.

In addition to noble gases, noble fission product metals like niobium, molybdenum, ruthenium, antimony and tellurium, which did not form stable fluorides in normal environment in the MSRE accompanied then xenon and krypton in the purge stream. The bulk of the fission product elements (lanthanides) form stable and soluble fluorides and remained with the fuel salt.

Fused salt volatilization technique based on bubbling of fluorine gas into fused fluoride salt containing elements forming volatile fluorides was used to recover uranium as volatile UF_6 from the molten fluoride salt carrier (based on $LiF-BeF_2$ or $LiF-BeF_2-ZrF_4$ mixtures). Special equipment for continual fluorination of the salt was designed and realized. The crucial part of the equipment was the continuous fluorination reactor with frozen wall for corrosion protection. The carrier molten salt containing dissolved UF_4 flowed into the top of the fluorination and was contacted by a counter current stream of fluorine gas, which stripped out the uranium in the form of volatile UF_6 .

Extraction of protactinium and rare earth elements

The main principles of the radiochemical technology were based on selective molten salt/liquid metal reductive extraction into liquid bismuth in multi-stage counter-current extraction system. Extraction to the liquid bismuth was proposed due to its suitable properties. Bismuth has a low melting point ($271^\circ C$), negligible vapour pressure in the temperature of interest ($500-700^\circ C$) and good solubility of lithium, thorium, protactinium, uranium and lanthanides. Bismuth is also essentially immiscible with molten halides. Reductive extraction between metal in molten salt and liquid metal phases can be expressed by following general reaction:



in which a metal halide MX_n in the salt reacts with lithium from the bismuth phase to produce M in the bismuth phase and the respective lithium halide in the salt phase. This reprocessing concept of 1970s is still a respected reference flow-sheet for current research and development in the area of MSR including the MSR Gen IV systems.

TMSR reprocessing concept

The TMSR fuel salt is constituted of $LiF-ThF_4$ (and eventually NaF or CaF_2). The different steps can be described as:

Helium bubbling for extraction of Kr, Xe and of a large part of ZrF_4 . Ba, Sr and Cs daughter nuclides of Xe and Kr. Depending on the bubbling efficiency, they will be removed during this step. Moreover, the bubbling contributes to the extraction of the noble metals (under particles form in the molten salt) by a process close to the flotation one.

- a) Fluorination process for extraction of U, Np, Pu, Te, Nb, Ru, I_2 , Mo, Cr, Tc under gaseous states. UF_6 , PuF_6 and NpF_6 are, after reduction, re-introduced in the fuel salt.
- b) A first reductive extraction step in liquid Bi whose redox potential is controlled by a given quantity of metallic thorium (pool-1). The objective of

this step is to remove all actinides present in the fuel salt and to leave the lanthanides in the salt.

- c) A second reductive extraction step in a second pool of liquid Bi saturated with 90% of thorium (pool-2). Due to the very low redox potential obtained at the Bi interface in these conditions, the reduction of almost all lanthanides is expected.
- d) Anodic oxidation of actinides. The fuel salt after removing of actinides and lanthanides is contacted with the pool 1 for anodic oxidation of actinides. The reactions involved in this step are given in Table 11.
- e) Anodic oxidation of lanthanides in a chloride medium. The anodic oxidation of lanthanides is performed by contacting the pool 2 with the chloride medium LiCl-KCl.
- f) Oxide precipitation of lanthanides. This precipitation is realized by a H₂O/HCl gaseous mixture bubbling in the chloride molten salt. The oxides so precipitated are sent to a waste storage. HCl gas is used for chlorination of metallic Li obtained at the cathode during the anodic oxidation of lanthanides. The reaction between Li and HCl produces H₂ gas which is used to balance the excess of ThF₄ in the cleaned fuel salt.

Table 11 Chemical reaction involved in the reductive extraction steps

Actinide extraction	
Reductive oxidation by Th	$4/3\text{AcF}_3 + \text{Th}(\text{Bi}) = \text{ThF}_4 + 4/3 \text{Ac}(\text{Bi})$
Anodic oxidation of Ac(Bi)	
Anode	$\text{Ac}(\text{Bi}) + 3\text{F}^- = \text{AcF}_3 + 3\text{e}^-$
Cathode	$\text{ThF}_4 + 4\text{e}^- = \text{Th} + 4\text{F}^-$
Balance	$4/3\text{AcF}_3 + \text{Th}(\text{Bi}) + 4/3 \text{Ac}(\text{Bi}) + 4\text{F}^- + \text{ThF}_4 + 4\text{e}^- = \text{ThF}_4 + 4/3\text{Ac}(\text{Bi}) + 4/3 \text{AcF}_3 + 4\text{e}^- + \text{Th} + 4\text{F}^-$
Lanthanides extraction	
Reductive extraction by Th	
Anodic oxidation of Ln(Bi) in chloride media	
Anode	$\text{Ln}(\text{Bi}) + 3\text{Cl}^- = \text{LnCl}_3 + 3\text{e}^-$
Cathode	$\text{LiCl} + \text{e}^- = \text{Li} + \text{Cl}^-$
Lanthanide oxide precipitation	$2\text{LnCl}_3 + 3\text{H}_2\text{O} = \text{Ln}_2\text{O}_3 + 6\text{HCl}$
Lithium chlorination	$\text{Li} + \text{HCl} = \text{LiCl} + 1/2\text{H}_2$
Reduction of ThF ₄ with H ₂	$\text{ThF}_4 + 2\text{H}_2 = \text{Th}(\text{Bi}) + 4\text{HF}$
Balance	$4/3\text{LnF}_3 + \text{Th}(\text{Bi}) + 4/3\text{Ln}(\text{Bi}) + 4\text{Cl}^- + 4\text{LiCl} + 4\text{e}^- + 4/3\text{LnCl}_3 + 2\text{H}_2\text{O} + 4\text{Li} + 4\text{HCl} + \text{ThF}_4 + 2\text{H}_2 =$ $\text{ThF}_4 + 4/3\text{Ln}(\text{Bi}) + 4/3\text{LnCl}_3 + 4\text{e}^- + 4\text{Li} + 4\text{Cl}^- + 2/3\text{Ln}_2\text{O}_3 + 4\text{HCl} + 4\text{LiCl} + 2\text{H}_2 + \text{Th}(\text{Bi}) + 4\text{HF}$ $4/3\text{LnF}_3 + 2\text{H}_2\text{O} = 2/3 \text{Ln}_2\text{O}_3 + 4\text{HF}$

Summary on MSR carrier molten salt from the viewpoint of fuel salt processing

The development of on-line reprocessing (fuel salt clean-up) technology represents a very specific problem affecting even the MSR design, reactor core chemistry and the choice of structural materials. Particularly, the link to reactor core chemistry is close because the chemical reactions rates and their character in the reactor have to be compensated by the reprocessing technology. A typical example is that nuclear fission in the molten salt fuel medium is an oxidizing process, nevertheless the redox potential in the reactor must be kept in slightly reductive range to protect the reactor core and primary circuit structural materials (nickel alloys and graphite). Therefore a very special attention should be paid to the selection of carrier molten salt, which must exhibit several basic properties (e.g. good thermal conductivity, appropriate melting point, low vapour pressure, radiation stability, sufficient solubility of actinides and last but not least the reprocessability by adequate separation techniques). Based on these requirements, the ${}^7\text{LiF}\text{--}\text{BeF}_2$ eutectics still remains the basic carrier salt candidate among several others, but the new studies enlarged the candidate carriers for some new ones.

- a) ${}^7\text{LiF}\text{--}\text{BeF}_2$ eutectics (FLiBe) – The salt is still a reference, originally proposed for MSBR. It has acceptable melting point, good solubility for uranium and thorium fluorides. However the thermodynamical stability of BeF_2 seems to be insufficient for electrochemical separation and also the solubility of transuranium fluorides is limited. On the other side, $\text{LiF}\text{--}\text{BeF}_2$ is immiscible with some liquid metals like bismuth and therefore it can be excellently used in molten salt/liquid metal reductive extraction process. In this process, a thermodynamical stability of the salt was testified. The molten salt/liquid metal reductive extraction technology was significantly verified in laboratory conditions during the MSRE and MSBR programs in 1960s and 70s.
- b) ${}^7\text{LiF}\text{--}\text{BeF}_2$ with addition of NaF – The salt should show an increase in solubility of transuranium elements, therefore it is often proposed for MSR actinide burners.
- c) ${}^7\text{LiF}\text{--}\text{ThF}_4$ – The salt is proposed for TMSR. The reprocessability of the salt has to be verified. The advantage of the $\text{LiF}\text{--}\text{ThF}_4$ mixture should be in elimination of problems with low BeF_2 thermochemical stability, the drawback could be in the significantly higher melting point.
- d) Fluoride salts containing ZrF_4 , RbF and other fluorides – Reprocessability of these salts by suitable pyrochemical technologies seems to be either not known (RbF) or problematic (ZrF_4), the proposals usually come from the wish to eliminate beryllium-containing salts due to toxicity of beryllium or

eliminate the need of use of expensive ^7Li isotope in LiF or to increase solubility of AnF_3 in the solvent system.

- e) Use of LiF-NaF-KF , LiF-NaF , LiF-CaF_2 and chloride systems – These salts could be satisfactorily used for the individual electrochemical separation processes within the reprocessing technology if actinides and fission products are completely extracted from the fuel salt carrier and subsequently dissolved in molten salts mentioned above.

Successful solution of the MSR fuel salt clean-up development is one of crucial steps before industrial deployment of MSR systems. As the MSR reprocessing technology must meet special demands (like radiation resistance, compactness, exclusion of moderating agents, compatibility with the carrier molten salt type and with the structural materials of MSR primary circuit, acceptable process reaction rate and process workability by remote handling), the pyrochemical separation processes, including the molten salt separation techniques, seem to be the only technologies which can be generally applied. The selection of molten salts used for these technologies will require still broad and intensive research and development activities and careful verification experiments. Also, the waste management will have to be solved in connection to the MSR fuel reprocessing development.

Summary

Different applications of molten salts in nuclear systems have been identified, for fuel salts and coolant salts.

The $^7\text{LiF-BeF}_2$ (66:34 in mol%) salt is the selected fuel carrier for the moderated (thermal) molten salt thorium breeder, giving as fuel salt $\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$. From neutronic as well as chemical point of view, there are no alternatives for this salt that do not penalise the breeding capacity of the reactor.

$^7\text{LiF-ThF}_4$ (78:22 or even 71:29 in mol%) is the reference fuel solvent composition for the non-moderated (fast) molten salt thorium breeder reactor. The neutronic analysis of the TMSR concept has demonstrated the feasibility of the concept, but it must still be clarified whether the physico-chemical properties (melting temperatures, solubility for the actinide trifluorides, density, expansivity, viscosity, thermal conductivity, heat capacity) of this salt fuelled by significant amount of UF_4 (2-4% of the total heavy nuclei in the moderated and 12-18% in the non-moderated systems) or AnF_3 (up to 25% of the total heavy nuclei in the non-moderated concept) are consistent with safe operation of the reactor and fuel salt clean-up unit. To tune these properties, addition of other components is possible. The most obvious is BeF_2 but there is an incentive to keep the content of this material low (e.g. $71\text{LiF-2BeF}_2\text{-27ThF}_4$ or $75\text{LiF-5BeF}_2\text{-20ThF}_4$ in mole%) or even zero. Alternatives are NaF and possibly CaF_2 . Therefore, the $^7\text{LiF-NaF-ThF}_4$ system must be further analysed, whereas

scoping studies of the ${}^7\text{LiF-CaF}_2\text{-ThF}_4$ system are required to assess the feasibility of this composition, including suitability for fuel salt processing.

The molten salt actinide burner is a non-moderated system also. The carrier salt for this application must have good solubility for the actinide trifluorides and this can be achieved using ${}^7\text{LiF-NaF}$ as solvent or ${}^7\text{LiF-(NaF)-BeF}_2$. Again, the goal is to keep the content of BeF_2 low or even zero. An interesting alternative is the use of plutonium and minor actinides as start-up for the thorium cycle in the MSR, leading to ${}^7\text{LiF-NaF-ThF}_4$ carrier salt.

In summary, it is clear that the ${}^7\text{LiF-(NaF)-AnF}_4\text{-AnF}_3$ salt (where An represent actinides) is the key system to be investigated in parallel to the ${}^7\text{LiF-(NaF)-BeF}_2\text{-AnF}_4\text{-AnF}_3$ system. Optimisation of the fractions of the components is still needed with respect to mentioned-above physico-chemical properties, corrosion behaviour in the Ni-Mo alloys and fuel salt processing.

For coolant salts, one has to make a distinction between salt for in-core use (primary coolant) and salts for out-of-core use (secondary or intermediate coolants). For primary coolants in thermal reactors, the requirements are very similar to thermal breeder reactors and ${}^7\text{LiF-BeF}_2$ (66-34 with $T_m=460^\circ\text{C}$) is the main candidate, with ${}^7\text{LiF-NaF-KF}$ (46-11.5-42.5 with $T_m=454^\circ\text{C}$), ${}^7\text{LiF-NaF-RbF}$ (46.5-6.5-47 with $T_m=426^\circ\text{C}$) and ${}^7\text{LiF-NaF-BeF}_2$ (30.5-31-38.5 with $T_m=316^\circ\text{C}$) as alternatives. Note that the last alternative molten salt mixture has the lowest liquidus temperature.

For secondary coolant applications, neutronic considerations nor actinide solubility play a role and a wider choice of materials is possible. For MSRs in which tritium control is the main concern, the NaF-NaBF_4 (8:92 with $T_m=385^\circ\text{C}$) system is the prime candidate, mainly because of its satisfactory tritium trapping. A ternary salt LiF-NaF-BeF_2 should be considered in future studies as alternative secondary coolant because a freezing temperature range of about $315\text{-}335^\circ\text{C}$ would be a practical value for engineering consideration.

For solid fuel fast reactors, no salt has been clearly identified as prime candidate in this study. However, recent studies at MIT have identified NaCl-KCl-MgCl_2 as a promising candidate.

Finally, heat transfer applications require a cheap and stable salt. $\text{NaNO}_3\text{-KNO}_3$ possibly with addition of NaNO_2 is the main candidate identified at this stage.

The salt properties are very important for the assessment of the feasibility of the various nuclear applications of molten salts. As discussed, a distinction must be made for fuel salts and coolant salts, although many common aspects can be identified. In general one can group the properties in (a) melting temperature, (b) physico-chemical properties, and (c) actinide solubility.

The melting temperatures of most binary and ternary salts are reasonably well known, and reliable thermodynamic models have been developed to predict melting points of ternary and quaternary mixtures.

Exceptions are salts containing significant amount of transuranium actinides of relevance to burner reactors. In this field, a significant effort is needed in the near future. The main needs are thus (i) measurements on plutonium and other actinide trifluorides (ii) verification measurements in ternary and quaternary systems to check and improve thermodynamic models.

Physico-chemical properties (density, viscosity, heat capacity, thermal conductivity) are poorly known for most of the salts that have been identified. An exception is the $7\text{LiF}-\text{BeF}_2-\text{ThF}_4-\text{UF}_4$ system that was extensively studied in the 1960s. Of these properties, the density (or molar volume) follows ideal behaviour and can be easily obtained from the pure compounds. This is not true for the other properties, but there is generally lack of data. Systematic experimental studies are needed here, especially on composition with actinides. Especially thermal conductivity is poorly known, and also theoretical models are poorly developed. In this field, the benefits from atomistic calculations must be explored, in combination with local structure determination techniques (NMR, Raman, EXAFS) to create the fundamental basis for the understanding of the molten salt properties.

Actinide solubility is a key issue for transmutation or burner fuels, but only a limited number of studies on PuF_3 solubility exist and none on the solubility of NpF_4 / NpF_3 , AmF_3 or CmF_3 . Solubility determinations in the systems with proper redox potential are therefore of prime importance. A prerequisite for such measurements is the availability of the pure actinide tri- and tetrafluorides in significant quantities, for which dedicated synthesis facilities are needed.

Conclusions

In a broader sense, the development and commercialization of a new coolant technology and its use in multiple reactor concepts is a major challenge. However, the increasing demands for high-temperature heat, the ability to convert such heat efficiently to electricity, and the interests in advanced reactors for breeding and waste management are creating the incentives to develop this family of technologies. The development and commercialization of such a coolant technology opens several new frontiers for nuclear energy.

MSR concepts (TMSR) offer alternative options for breeding and waste reduction with the added value of liquid fuel (intrinsic safety features, fuel cycle flexibility, on-line fuel processing, no fuel refabrication required, in-service inspection). For breeding, MSR is especially well fitted to thorium cycle.

The newly developed TMSR-NM concepts combine the assets of high temperature operation (cf. VHTR), breeding capability together with good intrinsic safety parameters in fast spectrum (unique among fast spectrum systems, SFR, LFR), high efficiency for waste minimization or reduction, and competitive economics. Attractive MSR designs are available for breeding (TMSR in thermal

and fast spectrum) and for MA burning (MOSART). They are robust reference configurations (with significant improvement compared to MSBR), allowing to concentrate on remaining R&D issues.

The development of liquid salts is significantly impeded by the complexity of their behaviour, as a result of their multi-component nature, in contrast to mono-specific coolants. This is a long term perspective to understand the behaviour and properties of liquid salts and to proceed towards a predictive approach making easier the selection of the appropriate salt composition for every given application.

Physics of Molten Salt Reactors

Physics aspects of TMSR investigated

- (i) influence of the chemical reprocessing on the neutronic behavior
- (ii) burning capabilities
- (iii) deployment capabilities, and
- (iv) deterministic safety analysis

MCNP neutron transport code and a materials evolution code REM were employed for this purpose. The former evaluates the neutron flux and the reaction rates in all the cells while the latter solves the Bateman equations for the evolution of the materials composition within the cells. These calculations take into account the input parameters (power released, criticality level, chemistry), by continuously adjusting the neutron flux or the materials composition of the core. These codes have been applied first to a general thorium molten salt reactor (TMSR) concept and then to a non-moderated TMSR.

General Thorium Molten Salt Reactor (TMSR) Concept

The general Thorium Molten Salt Reactor (TMSR) concept is a 2500 MWth (1 GWe) graphite moderated reactor based on the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle. The graphite matrix comprises a lattice of hexagonal elements with 15 cm sides. The density of this nuclear grade graphite is set to 1.86. The salt runs through the middle of each of the elements. For the systematic studies presented in this section, the salt considered in the simulations was a binary salt, $\text{LiF} - (\text{HN})\text{F}_4$, whose $(\text{HN})\text{F}_4$ proportion is set at 22 mole % (eutectic point), corresponding to a melting temperature of 565°C.

A graphite radial blanket containing a fertile salt (LiF 78 mole% - ThF_4) surrounds the core to protect the external structures, so that approximately 80 % of the escaping neutrons are stopped, and the breeding is improved. As a first step in reprocessing, helium bubbling in the salt circuit extracts the gaseous fission products and the noble metals within 30 seconds. To optimise the extraction efficiency which depends on the interaction between the liquid salt, the metallic clusters and the gas bubbles, scientific investigations and dedicated measurements are necessary. Studies are also needed to determine how to separate the fission products from the gas, store them and purify the gas. A delayed reprocessing of the total salt volume over a 6 month period with complete extraction of the fission products and of the transuranic elements is the second step. A fraction of salt is periodically set aside to be reprocessed off-line (batch mode). The fissile matter (uranium) can be extracted quickly by fluorination and sent back into the core. The other actinides and lanthanides can be separated via

several methods like electrolysis, reduction into metallic solvents, solid precipitation, or any other method developed in the frame of pyrochemistry reprocessing. Finally the actinides are sent back into the reactor core to be burnt, while the lanthanides are stored. The performance of the reactor, in terms of breeding and deployment capabilities, depends directly on the rate at which this off-line reprocessing is done. The ^{233}U produced in the blanket is assumed to be extracted within a 6 month period.

The moderation ratio can be altered by changing the channel radius. This modifies the neutron spectrum of the core, placing it anywhere between a much thermalized neutron spectrum and a relatively fast spectrum. The core size is adjusted to keep the whole salt volume constant and equal to 20 m^3 . Figure 1 shows the influence of the channel radius on the neutronic behaviour. In order to evaluate the performance of a reactor configuration, a number of parameters are checked: total reactivity feedback coefficient, breeding ratio, graphite life span and initial fissile inventory.

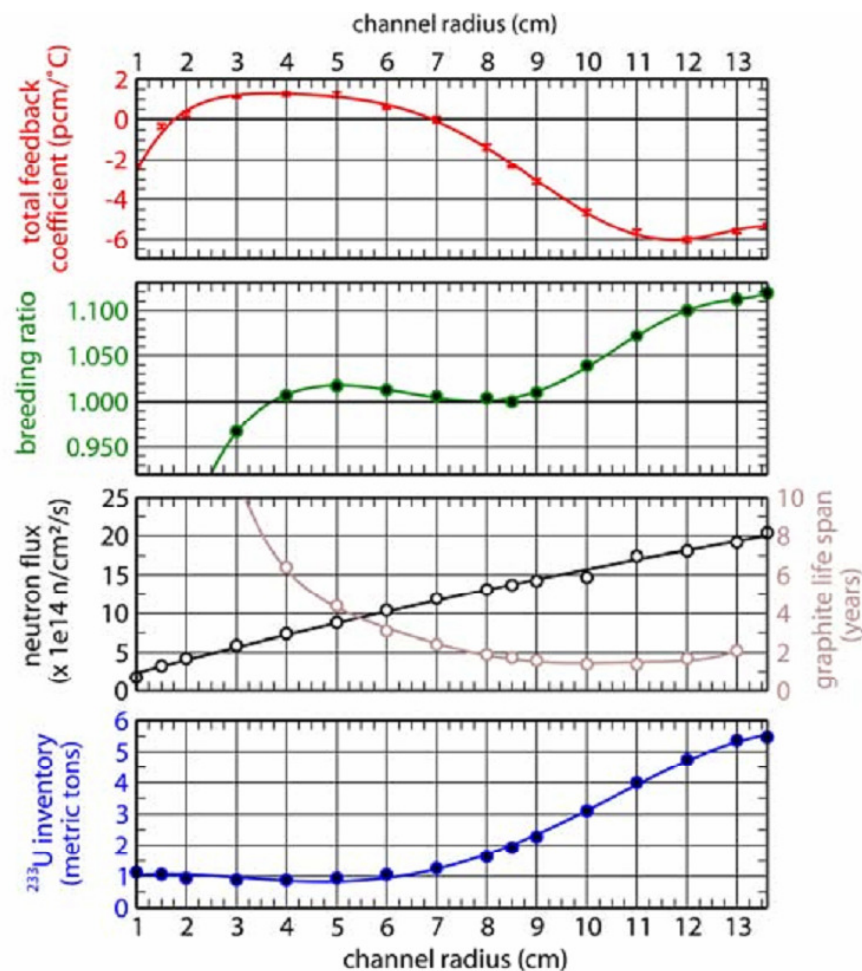


Figure 1 Influence of the channel radius on the TMSR's behaviour. Note: There is no graphite life span point for the 13.6 cm radius since there is no more graphite in the core

A wide variety of neutronic behaviours is available by changing the moderation ratio: thermal, epithermal and fast spectrum. A thermal spectrum leads to a low fissile inventory but slightly positive feedback coefficient due to the graphite, an epithermal spectrum like that of the MSBR requires efficient salt processing and leads to safety coefficients that are too limited while a fast spectrum implies a high breeding ratio and excellent safety coefficients but requires a large fissile inventory. Extensive studies have been carried out on these three fields of research to improve these configurations and to find relevant solutions. In this paper, the configuration without any graphite moderator in the core is considered, since it appears to be the most promising in terms of safety coefficients, reprocessing requirements, breeding and deployment capabilities.

The Non-Moderated Thorium Molten Salt Reactor

As shown in Figure 2, the core is now a single cylinder (1.25 m radius and 2.60 m height) where the nuclear reactions occur within the flowing salt. One third of the 20 m³ of fuel salt circulates outside of the neutron flux in external circuits flowing through the pumps, the gaseous extraction system which removes the gaseous and non-soluble fission products and the heat exchangers before coming back into the core. A fertile blanket surrounds the core.

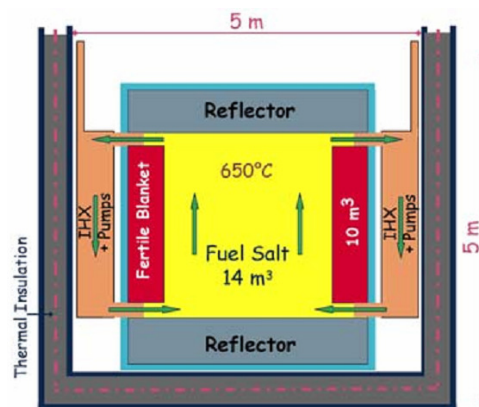


Figure 2 To scale vertical representation of the TMSR, including pumps and intermediate heat exchangers (IHX)

Fuel Salt Composition

The core contains a fluoride fuel salt, composed of LiF enriched in ⁷Li (99.999 %) and heavy nuclei (HN) amongst which the fissile element, ²³³U or Pu.

Parametric studies of this salt composition by varying the proportion of heavy nuclei in the fuel salt, indicated their influence on neutron energy moderation, actinide solubility, and the initial fuel inventory. For HN proportions ranging from 20 to 30 mole%, a binary salt LiF-(HN)F₄ has been chosen whose melting point is around 570°C. For lower proportions of HN, the calculations have been done with a salt containing 80 mole% of LiF completed with BeF₂ to lower the eutectic point temperature and to allow operation at 630°C. The salt

density ranges from 3.1 to 4.6 according to the HN proportion, with a dilatation coefficient of $10^{-3}/^{\circ}\text{C}$.

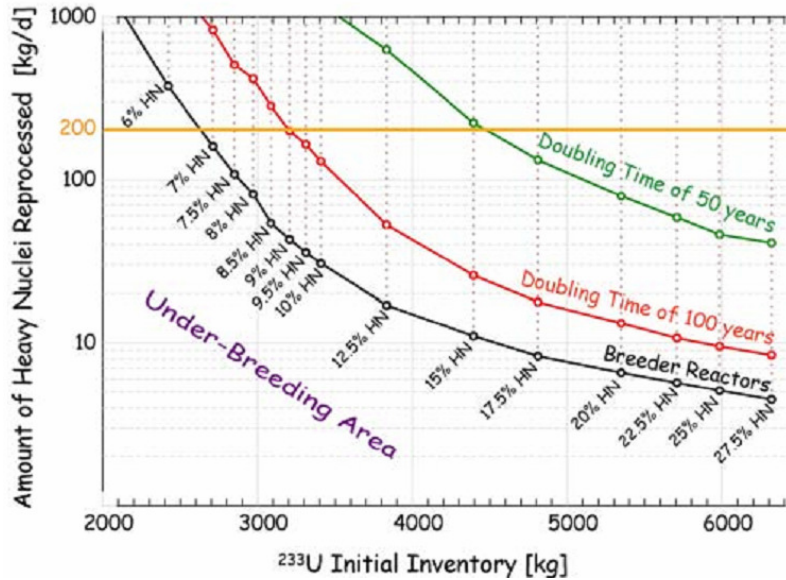


Figure 3 Amount of heavy nuclei reprocessed per day versus initial fissile (^{233}U) inventory, for different heavy nuclei proportions in the fuel

The results of these parametric studies are presented in Figure 3 where the reprocessing capacity required for a TMSR system is shown. The ordinate is the weight of heavy nuclei reprocessed per day while the abscissa corresponds to the initial fissile (^{233}U) inventory. Heavy nuclei proportions in the MSR fuel are indicated (x %mole HN). For each proportion of heavy nuclei in the salt, ranging from 6 mole % to 27.5 mole %, the breeding ratio of each reactor configuration as a function of the amount of heavy nuclei reprocessed per day is evaluated by simulation. The black line separates breeding and non-breeding zones. In the breeding zone, the red and green lines indicate the reprocessing requirements which allow the generation of the ^{233}U needed for a new TMSR reactor with the same inventory over 100 years and 50 years respectively. The ^{233}U initial inventory ranges from 2400 kg for a HN proportion in the salt of 6 mole % to 6300 kg for a HN proportion of 27.5 mole %. This corresponds to a variation of the neutron spectrum from an epithermal to a fast spectrum, as shown in Figure 4.

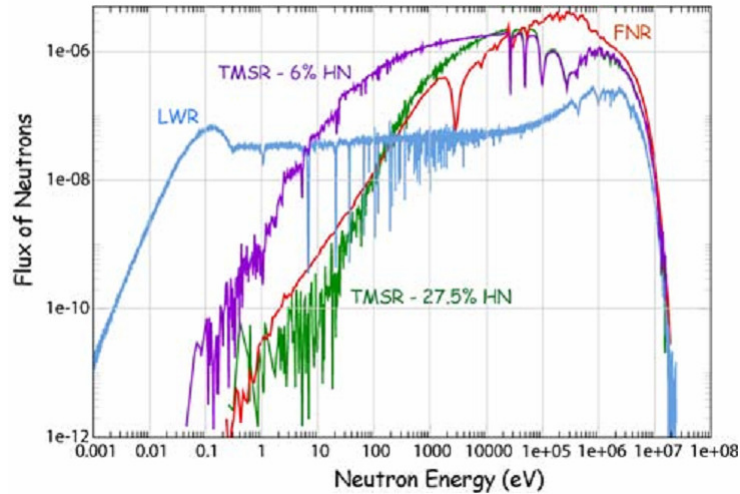


Figure 4 Neutron spectrum of two TMSR configurations (6% and 27.5% of heavy nuclei in the salt) compared to the neutron spectrum in a Light Water cooled Reactor (LWR) and in a Fast Neutron Reactor (FNR)

Heavy Nuclei Inventory and Burning Capabilities

Figure 5 illustrates the evolution of a typical fuel salt composition (17.5% mole HN) all along the operation of this reactor, for the ^{233}U -started (solid lines) and for the transuranic-started (dashed lines) TMSR.

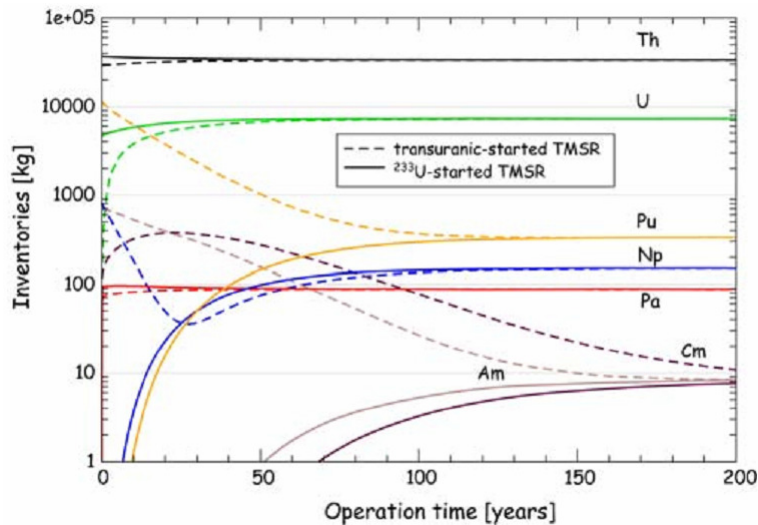


Figure 5 Heavy nuclei inventory for the ^{233}U -started TMSR (solid lines) and for the transuranic-started TMSR (dashed lines) with 17.5 mole% of heavy nuclei in the salt

In terms of transuranic inventory, as shown in Figure 5, the TMSRs started with transuranic elements tend towards TMSRs directly started and operated with ^{233}U after about forty years for a fuel salt with 17.5% of heavy nuclei, where more than

85% of the initial TRU inventories are burned. More generally, the assets of the Thorium fuel cycle are finally recovered for these transuranic-started TMSRs after 25 to 50 years for HN proportions ranging from 7 to 27.5 mole %, except for minor actinides where longer times are needed.

Deployment Capabilities

The deployment capabilities of a fleet of 4th generation reactors are directly linked to the breeding capabilities of each reactor since their initial fissile matter (^{233}U or Pu) does not exist on earth. The interesting variable is thus the operating time necessary to produce one initial fissile (^{233}U) inventory, called the reactor doubling time. the amount of excess ^{233}U produced in each TMSR configuration was studied. Transuranic-started TMSRs allow the extraction of significantly larger amounts of ^{233}U during their first 20 years of operation, thanks to the burning of TRUs which saves a part of the ^{233}U produced in the core. The higher deployment capabilities allowed by the use of TRUs in the transuranic-started TMSR are visible on the reactor doubling times, displayed in Figure 6 (dashed line), where the configurations with HN proportions larger than 15% have the lower reactor doubling times, around 30-35 years, to be compared with the doubling times greater than 45-50 years reachable with the ^{233}U -started TMSRs.

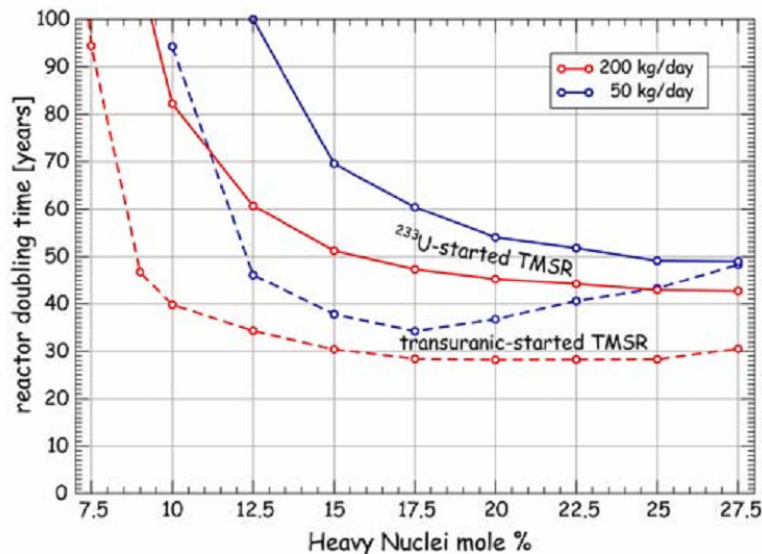


Figure 6 Reactor doubling time of ^{233}U -started (solid lines) and transuranic-started TMSRs (dashed lines) for the different HN proportion configurations and for two off-line reprocessing schemes: 200 kg (red lines) and 50 kg (blue lines) of HN reprocessed per day

Safety considerations

Chemistry related safety considerations

Safety aspects have been rather poorly addressed in recent MSR projects. However, since the premature termination of the MSBR program in 1975, safety rules for nuclear reactors have changed and the analysis performed in the 70's is unlikely to have the adequate or comprehensive level that would be required for the licensing of MSR today. Some safety aspects more specific to the choice of multi-component liquid salts are addressed: boiling temperature, vapour pressure, chemical reactivity of salts with other coolants.

Safety related issues and liquid/molten salts

A molten salt reactor can be operated with acceptable effect on the environment and safety of the public provided that the radioactive liquids and gases that circulate throughout much of the plant are managed carefully. Because the MSR processing plant is coupled directly to the reactor, there is no need for shipment of short-cooled fuel. Fission products extracted from the fuel salt are stored as solids or concentrated noble gases for shipment after long decay.

Production of tritium should be considered as a major issue for reactors where the salt composition includes LiF. It is necessary to use ^7Li because ^6Li has by order higher cross section for creation of tritium. Tritium that is produced in the fuel salt (from LiF component) in substantial quantities presents special containment problems. Much of the tritium is expected to diffuse through the walls of the tubes of the primary heat exchangers into the salt in the secondary system. Tritium can be extracted from the secondary salt and stored, but unless very special measures are taken, some tritium can be expected to diffuse into the steam system and be discharged from the plant. This steam blow down would be the only significantly radioactive effluent from the plant during normal operation. When the primary systems of an MSR are opened for maintenance, the copious amounts of fission products distributed throughout the systems require that stringent precautions be taken to prevent undesirable releases into the atmosphere. By use of evaporators and demineralizers to process wastes and the liquids that are used for decontaminating and washing radioactive equipment during maintenance, the radioactivity discharged in these liquids can be kept to insignificant amounts.

With regard to safety from severe accidents, the use of fluid fuel places some special requirements on the design and operation of an MSR. At the same time, it eases or eliminates some requirements that are important in solid fuel reactors (e.g. mitigation of severe accidents with core meltdown). Although it is clear a priori that different measures must be taken to ensure safety, whether these measures are more or less complex and expensive in an MSR than in other reactor systems can be determined only by a comprehensive analysis that begins with the most basic considerations.

Fuel salt homogeneity and safety

A unique consideration in fluid fuel reactors is the possibility of inhomogeneity of the fissile material in the circulating fuel. Specifically of concern is gradual segregation of fissile material outside the core, followed by rapid introduction with the incoming stream. The MSR fuel salt, is quite stable over a range of conditions much wider than the anticipated deviations. Segregation of uranium could conceivably be produced by introduction of reducing agents or oxygen into the salt, but adequate protection against this should be provided in the MSR (e.g. gettering action of the ZrF_4 for H_2O major impurity). The principal components of MSR fuel mixtures do not form intermediate compounds with PuF_3 . It is anticipated therefore that in concentrations at which PuF_3 would be used, it would not be deposited preferentially from the bulk salt during the inadvertent freezing, nor at locations such as in freeze valves.

Boiling temperature, vapour pressure and vapour species

Most fluoride salts exhibit very low vapour pressures. Only compounds with higher oxidation state cations (such as BF_3 , UF_6 , and MoF_6) exhibit high vapour pressures. A few of the elements useful for coolants (BeF_2 , ZrF_4) exhibit appreciable vapour pressures (> 1 mm Hg) at $800^\circ C$. However, mixtures of these pure components will always exhibit lower vapour pressures (higher boiling points) than the most volatile constituent. Therefore, these salts do not exert significant vapour pressures (> 1 bar) except at very extreme temperatures. However, other factors are important. Even in a low-pressure system, the magnitude and nature of vapour produced from the salt need to be evaluated. Experience with the ARE and the MSRE shows that very low salt vapour pressures (< 1 mm Hg) simplify the off-gas system design and that certain vapour species can present problems.

In any high-temperature salt system, a purged cover gas will be necessary. The transport of significant amounts of salt vapour in this cover

gas system can cause problems. In the operation of the ARE, it was found that the vapour over the ARE salt (53%NaF-41%ZrF₄-6%UF₄) was nearly pure ZrF₄. Because ZrF₄ sublimes rather than boils, ZrF₄ "snow" was found in the exhaust piping. The ZrF₄ was not returned to the salt reservoir by condensing as a liquid and draining back to the salt reservoir. Elaborate "snow traps" were designed to mitigate this problem, but it appears that a wise choice of salt composition can eliminate it completely.

The experience with the MSRE was quite different. The MSRE salts (65%LiF-29%BeF₂-5%ZrF₄-1%UF₄) exhibited very low vapour pressure, more than 100 times lower than the ARE salt. The vapour over the MSRE salt was also of a different character. This vapour contained both LiF and BeF₂ in a proportion that melted at a low temperature, such that the condensate would drain back to the reservoir as a liquid.

Vapour pressure is the physical property that is most sensitive to salt composition. Studies have been conducted to understand the effect of composition on the vapour pressure and vapour species of the thermodynamically non-ideal systems containing ZrF₄ and BeF₂. The results of these studies are useful for understanding and selecting the optimum coolant salt composition and are briefly reviewed in the following paragraphs.

The effect of salt composition on vapor pressure can be explained with the Lewis acid-base theory. The native volatility of compounds containing the "acidic" constituent (Zr⁴⁺, Be²⁺) can be suppressed by donation of fluoride anions from the "basic" alkali fluoride constituent. The product of this donation is a low-volatility coordination-complex that is an integral part of the molten salt solution. Not all the alkali fluorides are equal in their ability to donate fluoride anions for coordination compounds. The affinity of alkali cations for their own fluoride anion decreases with increasing atomic number; thus, the heavier alkali elements will more readily donate their fluoride anions. Therefore, heavier alkali are more effective in reducing the native volatility of the compounds containing the acidic species (Zr⁴⁺, Be²⁺).

The effect of salt composition on vapour pressure is readily apparent in the BeF₂ and ZrF₄ systems, with suppression of volatility as the ratio of alkali fluoride content increases so that it satisfies the coordination-bonding demands of the polyvalent cation. The heavier alkali fluorides are more effective in suppressing the native volatility of the compound containing the polyvalent element (e.g., beryllium or zirconium).

The decrease of vapour pressure due to coordination bonding is also accompanied by a change in vapour composition. For a system rich in alkali fluoride, the vapour consists primarily of the alkali fluoride. For salt compositions that exist at the optimum ratio that just

satisfies the coordination demands of the system, the vapour species is an association complex of the alkali fluoride and the polyvalent cation. For systems deficient in alkali fluoride, the volatile species is the parent compound containing the polyvalent cation.

From a practical standpoint, one should favour salt compositions with very low vapour pressures (< 1 mm Hg at 900°C) that generate vapour species that readily melt after condensing. This corresponds to salt compositions with a ZrF_4 mole fraction in the range of ~ 20 – 45% , and with a mole fraction of BeF_2 less than ~ 35 – 45% , depending on the alkali cations present and the temperature under consideration.

Coolant salt interactions in MSR

From the safety standpoint, it is important that mixing of fuel and coolant salt, spillage of coolant salt into the containment cells, and leakage of steam into coolant salt should not give rise to situations that would endanger the health and safety of the public or of the operators of the plant. The only credible event that would produce mixing of fuel and coolant salts is a leak in a primary heat exchanger. No chemical reactions that would generate excessive heat or precipitate constituents of either salt would be expected to occur on mixing. Fuel salt and sodium fluoroborate are immiscible, however, so two salt phases would be present in both systems. Although the salts are immiscible, exchange occurs between the phases with lithium and beryllium fluorides entering the lighter fluoroborate phase and sodium fluoride and boron trifluoride moving into the fuel salt phase. Uranium and thorium fluorides remain in the heavy phase.

In the primary system, the exchange of constituents between salts would have no significant effect on the melting point of the fuel salt. The melting point of the coolant salt dispersed in the fuel would increase somewhat, some BF_3 would be released, and the BF_3 overpressure in the primary system would be expected to rise to about 5 atm. In the secondary system, the interaction between the fuel and coolant salts would tend to raise the liquidus temperature of the fuel containing salt but would not significantly affect the coolant salt. Since much of the secondary system normally operates at a temperature below the liquidus of the fuel salt, the fuel salt that leaked into the secondary system would initially be dispersed as frozen particles throughout much of one circuit. Whether the particles remained as solids would depend on measures taken to heat or cool the secondary circuit after the coolant salt had been drained.

None of the conditions associated with mixing of fuel and coolant salts in the primary or secondary systems appear to be capable of producing a

break in either system. The secondary circuits must be heavily shielded against the radioactivity present in the coolant salt during normal operation. This shielding can be made adequate to protect against the fission products that would be introduced by the fuel salt. Repairing or replacing the faulty heat exchanger, reclaiming the fuel salt, and disposing of the contaminated coolant salt promise to be unpleasant operations. The chemical toxicity of the borontrifluoride precludes its indiscriminate release from the plant, but the presence of sodium fluoroborate does not otherwise affect the safety of those operations.

Water and steam react with sodium fluoroborate to produce primarily hydrogen fluoride and sodium hydroxyl fluoroborate. The reactions are not destructively exothermic, but the hydrogen fluoride is corrosive to the metals of the reactor secondary system and the tubes that separate the fuel salt from the coolant salt. Although the corrosion rates are not catastrophic under any foreseeable circumstance, the leakage rate of water from the steam system into the secondary system and the hydrogen fluoride concentration in the secondary salt must be kept low in order to maintain a low corrosion rate of piping and equipment. In the event of a rupture of one or more tubes in a steam generator or superheater, the rapid pressurization of the secondary system and the possibility of transmitting that pressure to the primary system is the major concern. Isolation valves must be provided to stop the flow of feedwater and steam to the faulty steam generating equipment and pressure relief devices must be provided on the secondary system to keep the pressure below the system design pressure. The steam and salt that are discharged through these devices must be contained. The affected secondary system must be purged of hydrogen fluoride and moisture and the contaminated salt must be purified or replaced while repairs are made on the steam generator before operation of the plant can be resumed.

The use of a chemically reactive coolant in the secondary system of the MSR introduces some problems in designing the plant for upset conditions. The interactions of the coolant with the materials, with fluids in contiguous reactor systems, and with the cell atmospheres, however, do not appear to be so vigorous or the reaction products so aggressive as to create major safety concerns.

Physics aspects of Safety Considerations

The fraction of delayed neutrons, β , is very important in reactor control, it is a determining safety parameter. Seven precursor families listed in Table 1 for two fissile nuclei, ^{233}U and ^{235}U have been considered. The total number of fission product atoms giving rise to delayed neutron emissions will depend on the fissile composition of the reactor and,

to a lower extent, on the type of neutron spectrum. The value of β at equilibrium for the TMSR considered here has been estimated by simulation to 450 pcm.

Table 1 Abundances of seven delayed neutron precursors for two uranium isotopes

Group	1	2	3	4	5	6	7
Precursor	87Br	137I	88Br	93Rb	139I	91Br	96Rb
Half-Life	55.9 s	24.5 s	16.4 s	5.85 s	2.3 s	0.54 s	0.199 s
Abundances							
²³³ U (fast)	0.0788	0.1666	0.1153	0.1985	0.3522	0.0633	0.0253
²³³ U (thermal)	0.0787	0.1723	0.1355	0.1884	0.3435	0.0605	0.0211
²³⁵ U (fast)	0.0339	0.1458	0.0847	0.1665	0.4069	0.1278	0.0344
²³⁵ U (thermal)	0.0321	0.1616	0.0752	0.1815	0.3969	0.1257	0.0270
Mean Value	0.0742	0.1679	0.1209	0.1915	0.3533	0.0684	0.0240

Feedback Coefficient

The feedback coefficient or temperature coefficient is the variation of the multiplication coefficient dk for a given variation dT in temperature of the whole or part of the core. This feedback coefficient has to be negative to ensure the intrinsic stability of the reactor. The practical evaluation of a feedback coefficient is done as follows. The multiplication coefficient k is first computed for the core with the matter compositions at equilibrium for a temperature of 900 K. It is then re-calculated using the same compositions but at a different reactor temperature.

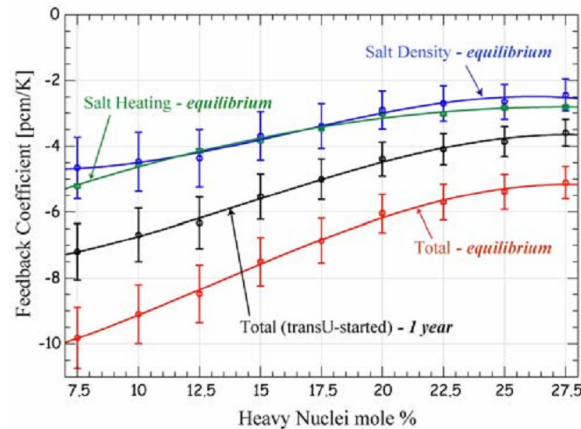


Figure 7 Feedback Coefficients of the TMSR started both with ²³³U or with transuranic elements at equilibrium and of the TMSR started with transuranic elements after one year of operation, as a function of the HN proportion in the fuel salt.

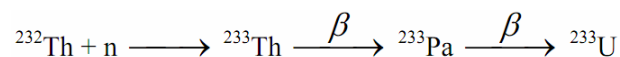
The total feedback coefficient at equilibrium is displayed in Figure 7, together with its components, the contributions of the salt heating and salt

density, as a function of the HN proportion in the salt. All these safety coefficients are significantly negative for all HN proportions, including the density coefficient which can be seen as a void coefficient. The total feedback coefficient is largely negative, ranging from -10 pcm/K to -5 pcm/K.

Reactivity Margins in the TMSR

Beyond the safety aspects, using a liquid fuel allows the adjustment of fertile and fissile matter without unloading the core, doing away with the need for any initial reactivity reserve, contrary to the case of a LWR where this reactivity reserve amounts to 10,000 pcm. Some reactivity margins may be introduced in the core of the TMSR involuntarily through four possible perturbations: a direct insertion of reactivity, the loss of salt circulation, loss of coolant, and finally the loss of the fertile blanket surrounding the core.

Unintentional introduction of ^{233}U in core: The reactivity increase of the TMSR is equal to 13.6 ± 0.2 pcm per extra kilogram of ^{233}U . For an iso-breeder reactor, the core needs to be fed 2.6 kg/day in ^{232}Th . If ^{233}U is added, instead of ^{232}Th , this would lead to an increase of the reactivity of 35 pcm/day. ^{233}U is also directly produced in core as follows:



The radioactive period of ^{233}Pa being 27 days, it decays rather quickly in core - around 2.55% per day - giving ^{233}U . This corresponds to (1) a disappearance of neutron capturing matter and (2) an appearance of fissile matter. If the reactor were to stop operating, the first effect would increase the reactivity by 33 ± 11 pcm/day, the second effect by 27 ± 1 pcm/day, giving a total reactivity margin of 60 ± 11 pcm/day.

Loss of salt circulation: As detailed earlier, the fraction of delayed neutrons for the whole salt at equilibrium is equal to 450 pcm. As two thirds of the salt is in the core and one third outside in the heat exchangers during normal operation, 2/3 of the delayed neutrons (300 pcm) are emitted within the core. If the salt circulation is stopped, all the delayed decays will occur in the reactor, since all the fission products will stay in core. This will represent an addition of 150 pcm to the multiplication coefficient in some ten seconds.

Loss of coolant: In TMSR case, the draining of the core is equivalent not only to a loss of the coolant but also to a loss of fuel, leading to a decrease of the

reactivity. Since the reactor is equipped with a draining system through stoppers made for example of solid salt and designed to melt at a given temperature, it was important to check that the salt evacuation will not worsen an accident, since it occurs mainly in abnormal situations.

Loss of fertilizer blanket: The structure of this blanket is made of ZrC, containing a salt composed of LiF (72.5%) – ThF₄ (27.5%). This salt may be solid or liquid. The second solution simplifies the recovery of the fissile matter but the blanket could then be accidentally drained. Simulations carried show that the extraction of the entire blanket leads to a decrease of reactivity. Because the ZrC structure of the blanket does not moderate neutrons, draining the blanket leads to increased neutron leakages and thus to a decrease of reactivity.

Table 2 Reactivity margins of a typical TMSR configuration

Origin	²³³ U Addition	²³³ Pa Decay	Salt Circulation	Core Draining	Blanket Draining	Reactivity Margins
Reactivity Margin	35 pcm	60 pcm	150 pcm	-	-	< 500 pcm
Time of Insertion	Some seconds	1 day	Some 10 s	-	-	> some seconds

Safety Evaluation of the TMSR

A single kinetic-point evaluation model was used for this purpose. Deterministic safety implies the definition of a viability domain, corresponding to the range of acceptable core parameters. As internal pressure is very low in a TMSR, only phenomena following a temperature increase of the fuel salt could endanger the reactor. As a consequence, the viability region is limited by the fuel solidification temperature with $T_{\min} = 800\text{K}$ as lower limit, and by the salt dissociation temperature with $T_{\max} = 1600\text{K}$ as upper limit.

This kinetic-point model assumes (i) A uniform distribution of the fissions within the core (correct in TMSR case), (ii) No heat propagation, and (iii) No follow-through of the precursors of the delayed neutrons.

Safety evaluation of the reference configuration

From Table 2, it can be concluded that the insertion of the total reactivity margins of the TMSR corresponds to the addition of less than 1000 pcm in some minutes (around 100 seconds). The insertion of this upper limit of 1000 pcm, in 0.001 to 100 seconds, is thus displayed on figure 7 in terms of temperature, power and reactivity evolutions. The calculations

have been done using a feedback coefficient equal to -5 pcm/K, close to the worst coefficient value. As expected, the final temperature reached at equilibrium is equal to $T_0 + \frac{1000\text{pcm}}{dk/dT} = 1100\text{ K}$. It can be concluded from

Figure 7 first that the prompt criticality is reached for an insertion time shorter than one second, since the maximal reactor reactivity is higher than the fraction of delayed neutrons (equal to 300 pcm). If, for example the reactivity insertion is expected within 0.001 second (figure 7, black curves), the evolution of the reactivity has to be considered first (figure 7, top) since it is modified directly. In this case, the whole 1000 pcm reactivity is added before the reactor begins to react. The reactivity insertion then leads to an increase of the reactor power (figure 7, middle) which reaches a maximum at 1 MW/cm³. Finally, the reactor temperature (figure 7, bottom) begins to increase after some milliseconds. A significant temperature increase triggers the reactor feedback, and as a consequence reactivity and power decrease. The heat in excess has finally to be evacuated to recover normal operating conditions.

However, even in the case of the prompt reactivity regime, a huge and dangerous increase of power and temperature could be avoided thanks to the reactor's safety parameters, as seen for the insertion time of 0.1 second. In this case (figure 6, green curves), the thermal feedback is fast enough to lower the reactivity back to zero before the end of the 1000 pcm reactivity insertion. A bounce can thus be observed, especially on the power evolution, when the remaining reactivity margins are inserted. A second feedback leads to the final reactivity and power decreases, and normal running conditions are reached again when the accumulated heat is evacuated.

For insertion times greater or equal to one second, and a fortiori in a more realistic case of 100 seconds, the reactor feedback is fast enough to avoid the prompt reactivity regime. Thus the reactor succeeds in absorbing at least a 1000 pcm reactivity insertion in one second and behaves safely.

Caution: The simulations carried out are reliable for the longer insertion times and are pessimistic for the shorter insertion times, since a uniform power distribution within the core is assumed and also time required to reach the density equilibrium (equilibrium considered as immediate). Larger local powers are indeed reached within the core, leading to a faster feedback to reactivity insertion than shown in figure 7. For the insertion times between 0.01 and 10 seconds, things are more complex and precise calculations have to be done mainly to take into account the follow-through of the precursors of the delayed neutrons.

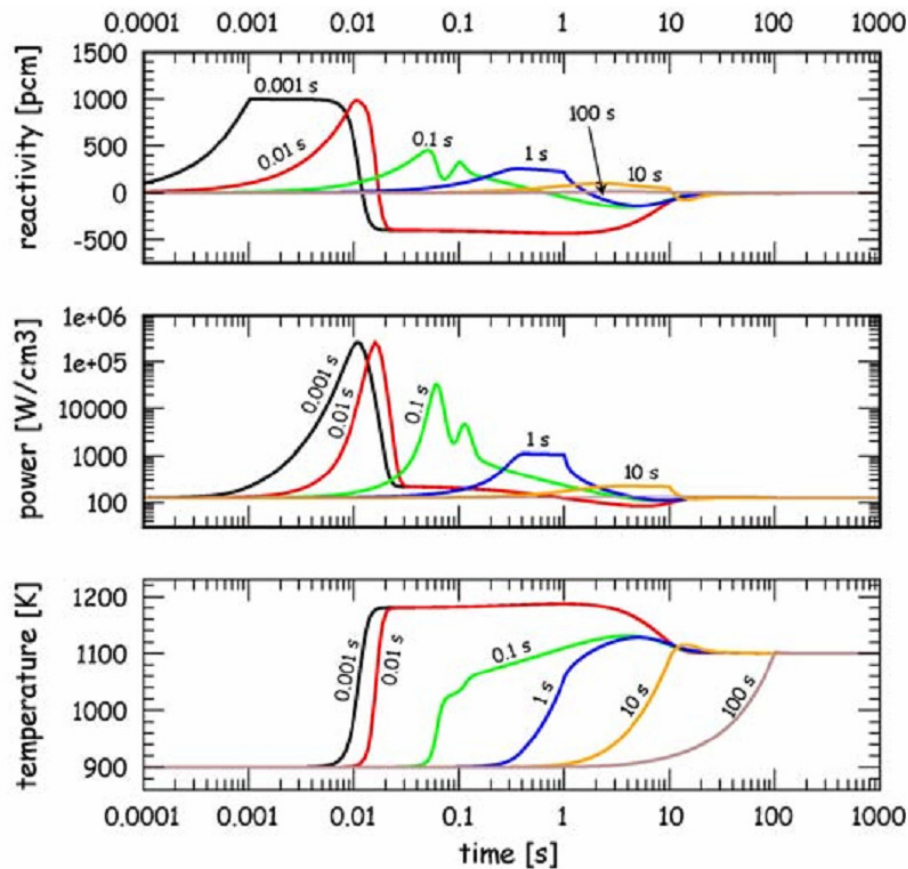


Figure 7 Evolution of the reactivity, the reactor's power and the salt temperature following an insertion of reactivity of 1000 pcm in 0.001 to 100 seconds, the total feedback coefficient of the reactor being equal to -5 pcm/K

Control of the reactor

The TMSR can be driven by the extracted power, and thus by the energy demand through the secondary circuit, as illustrated in Figure 8. As the energy demand increases, the power extracted becomes larger than the power produced by fission in the core. As a consequence, the temperature of the fuel salt decreases, leading to a reactivity increase as shown in Figure 8 for the extracted powers greater than 100% (red and green curves corresponding respectively to an extracted power of 125% and 150% of the nominal extracted power). This logically brings on a larger production of power in the reactor. This produced power then temporarily exceeds the power demand, which restores the initial temperature and reactivity. The reactor finally returns to equilibrium at its nominal temperature, while producing the needed power.

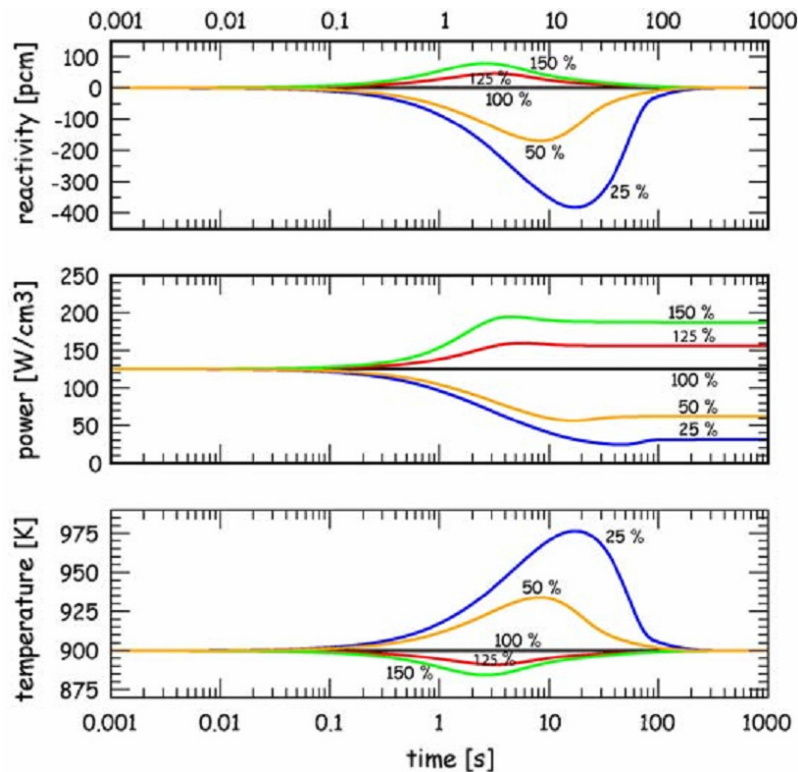


Figure 8 Evolution of the reactivity, the reactor's power and the salt temperature as a function of the extracted power (in percent of the nominal extracted power), for a total feedback coefficient of the TMSR equal to -5 pcm/K

As shown on the other curves of Figure 8 for extracted powers lower than 100% of the nominal reactor operation, the same goes in the case of a power demand decrease, this bringing on this time a rise of temperature, a drop of reactivity and thus a decrease of the produced power. The reactor's reactivity can also be controlled by stabilizing its operating temperature. A decrease of temperature with a constant produced power reveals a drop of reactivity due to a lack of fissile matter in the core. Reversely, an increased operating temperature reveals a too large reactivity which has to be corrected by stopping the addition of fissile matter (in the case of an under-breeder reactor for example).

The Advance High Temperature Reactor (AHTR)

The AHTR is a new reactor concept that has three technical characteristics: high temperature, passive safety, and a large power output. The high temperature is required to produce hydrogen and efficiently produce electricity. The passive safety features are required to reduce operating costs and improve public acceptance. The large power output, passive safety features, and high efficiency of electricity production (a consequence of high temperatures) are the enabling technologies to improve economics. The development is a joint effort of Oak Ridge National Laboratory, Sandia National Laboratories, and the University of California at Berkeley.

The AHTR (Figure 1 Table 1) uses coated-particle graphite-matrix fuels and a molten-fluoride-salt coolant. The fuel is the same type that is used in modular high-temperature gas-cooled reactors (MHTGRs), with fuel failure temperatures in excess of 1600°C . The optically transparent molten salt coolant is a mixture of fluoride salts with freezing points near 400°C and atmospheric boiling points of $\sim 1400^{\circ}\text{C}$. Several different salts are being evaluated as the primary coolant, including lithium-beryllium and sodium-zirconium fluoride salts. The reactor operates at near-atmospheric pressure. At operating conditions, the molten-salt heat-transfer properties are similar to those of water. Heat is transferred from the reactor core by the primary molten-salt coolant to an intermediate heat-transfer loop. The intermediate heat-transfer loop uses a secondary molten-salt coolant to move the heat to the turbine hall. In the turbine hall, the heat is transferred to a multi-reheat nitrogen or helium Brayton cycle power conversion system. For hydrogen production, the heat is transferred to the thermochemical hydrogen production facility, which converts water and high temperature heat to hydrogen and oxygen.

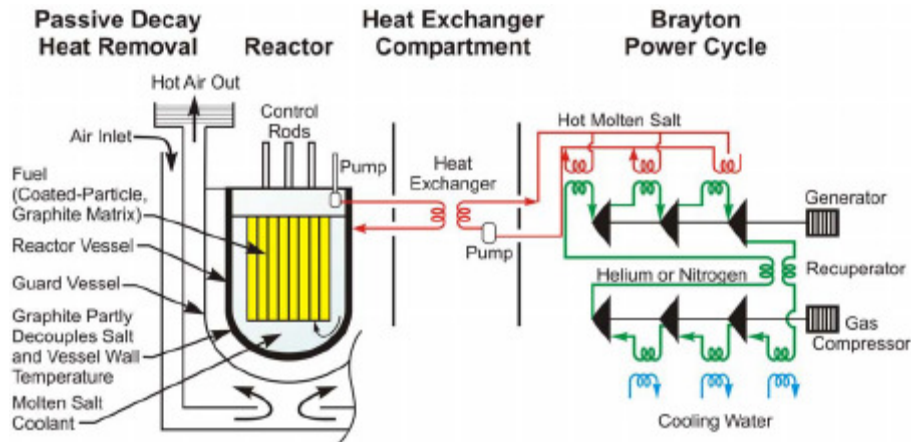


Figure 1 Schematic of the AHTR for electricity production

Table 1. AHTR preconceptual design parameters

Power level	2400 MW(t)	Power cycle	3-stage multi-reheat Brayton
Core inlet/outlet temperatures (options)	900°C/1000°C 700°C/800°C 670°C/705°C	Electricity (output at different peak coolant temperatures)	1357 MW(e) at 1000°C 1235 MW(e) at 800°C 1151 MW(e) at 705°C
Coolant	2^7LiF-BeF_2 (NaF-ZrF ₄)	Power cycle working fluid	Nitrogen (Helium: long term option)
Fuel		Vessel	
Kernel	U-carbide/oxide	Diameter	9.2 m
Enrichment	10.36 wt% ^{235}U	Height	19.5 m
Form	Prismatic	Reactor core	
Block diameter	0.36 m (across flats)	Shape	Annular
Block height	0.79 m	Diameter	7.8 m
Columns	324	Height	7.9 m
Decay heat system	Air cooled	Fuel annulus	2.3 m
Volumetric flow rate	5.54 m ³ /s	Power density	8.3 W/cm ³
Coolant velocity	2.32 m/s	Reflector (outer)	138 fuel columns
		Reflector (inner)	55 fuel columns

The baseline AHTR facility layout (Figure 2) that was developed is similar to the S-PRISM sodium-cooled fast reactor designed by General Electric. Both reactors operate at low pressure and high temperature; thus, they have similar design constraints. The 9.2-m-diam. vessel is the same size as that used by the S-PRISM. In the initial baseline studies, it was assumed that the fuel and power density (8.3 W/cm³) were essentially identical to those of the MHTGR. The better heat transfer of the molten salt relative to helium implies (1) lower peak reactor temperatures if the power densities are similar or (2) a higher-power-density core that is smaller or has a higher total power output for a given reactor vessel size. Three peak coolant temperatures are being considered: 705, 800, and 1000°C for the AHTR—Low Temperature (AHTR-LT), the AHTR—Intermediate Temperature (AHTR-IT), and the AHTR—High Temperature [AHTR-HT], respectively. The AHTR-LT uses existing materials, the AHTR-IT uses existing materials that have not been fully tested, and the AHTR-HT uses advanced materials. The AHTR-HT and AHTR-IT include a graphite blanket system, while the AHTR-LT has a metallic blanket system that separates and insulates the reactor vessel from the reactor core so that the fuel and coolant can operate at higher temperatures than the vessel. This insulation ensures long vessel life

(minimizing long term creep) and minimizes heat losses during normal operations.

In the proposed design, the AHTR has an annular core through which the coolant flows downward. The molten salt coolant flows upward through the non-fuel graphite section in the middle of the reactor. The molten-salt-coolant pumps and their intakes are located above the reactor core with appropriate siphon breakers; thus, the reactor cannot lose its coolant except by failure of the primary vessel. The guard vessel is sized so that even if the primary vessel fails, the core remains covered with salt.

Decay Heat Cooling and Accident Management

When a reactor shuts down, radioactive decay heat continues to be generated in the reactor core at a rate that decreases over time. The AHTR uses passive reactor vessel auxiliary cooling (RVAC) systems similar to that developed for decay heat removal in the General Electric sodium-cooled S-PRISM. The reactor and decay-heat-cooling system are located in a below-grade silo (Figure 1). In this pool reactor, RVAC system decay heat is (1) transferred from the reactor core to the reactor vessel graphite reflector by natural circulation of the molten salts, (2) conducted through the graphite reflector and reactor vessel wall, (3) transferred across an argon gap by radiation to a guard vessel, (4) conducted through the guard vessel, and then (5) removed from outside of the guard vessel by natural circulation of ambient air.

The rate of heat removal is controlled primarily by the radiative heat transfer through the argon gas from the reactor vessel to the guard vessel. Radiative heat transfer increases by the temperature to the fourth power (T^4); thus, a small rise in the reactor vessel temperature (as would occur upon the loss of normal decay-heat-removal systems) greatly increases heat transfer out of the system. The effective thermal inertia, per unit volume of the reactor vessel, is much larger than that for gas-cooled reactors because of the high heat capacity of the molten salt and the low radial temperature gradient across the reactor core.

Under accident conditions such as a loss-of-forced-cooling accident, natural circulation flow of molten salt up the hot fuel channels in the core and down by the edge of the core rapidly results in a nearly isothermal core with about a 50°C temperature difference between the top and bottom plenums. For a typical simulation of the reactor with a nominal coolant exit temperature of 1000°C, the calculated peak fuel temperature in such an accident is ~1160°C, which will occur at ~30 hours with a peak reactor vessel temperature of ~750°C at ~45 hours. The average core temperature rises to approximately the same temperature as the hottest fuel during normal operations.

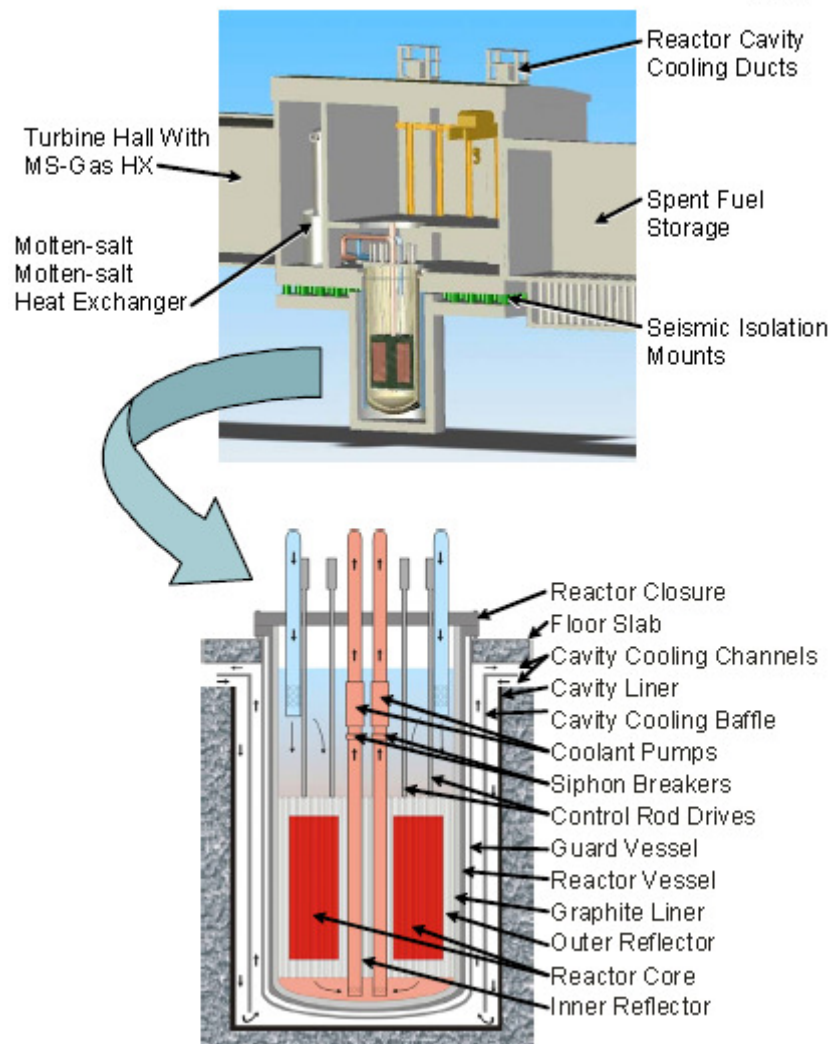


Figure 2 Schematic of the AHTR nuclear island and vessel

In terms of passive decay-heat-removal systems, a major difference is noted between the liquid-cooled AHTR and gas-cooled reactors. The AHTR can be built in very large sizes [>2400 MW(t)], while the maximum size of a gas-cooled reactor with passive-decay-heat removal systems is limited to ~ 600 MW(t). The controlling factor in decay heat removal is the ability to transport this heat from the center of the reactor core to the vessel wall or to a heat exchanger in the reactor vessel. The AHTR uses a liquid coolant where natural circulation can move very large quantities of decay heat to the vessel wall with a small coolant temperature difference ($\sim 50^{\circ}\text{C}$). Unfortunately, in a gas-cooled reactor under accident conditions when the reactor is depressurized, the natural circulation of gases is not very efficient to transport heat from the fuel in the center of the reactor to the reactor vessel. The heat must be conducted through the reactor fuel to the vessel wall. This

inefficient heat transport process limits the size of the reactor to ~600 MW(t) to ensure that the fuel in the hottest location in the reactor core does not overheat and fail.

In terms of various accidents, there are no major stored energy sources within containment such as high pressure fluids [helium and water] or reactive fluids [sodium]. This reduces requirements for the containment. The molten salt dissolves almost all actinides and fission products (except xenon and krypton). This provides a major barrier against the release of any radionuclide from fuel failure.

Reactor Physics and Fuel Cycle

Because the AHTR uses the same basic fuel type and the molten salt coolant has a low neutron-absorption cross section, the reactor core physics and fuel cycle options are generally similar to those for helium cooled high-temperature reactors. Reactor power is limited by a negative temperature coefficient, control rods, and other emergency shutdown systems.

Several molten fluoride salts with generally similar properties are being evaluated to determine the optimum coolant salt (Ingersoll et al. 2004). Such evaluations involve trade-offs in neutronics, cost, operations, and other parameters. The initial baseline AHTR design used the same salt ($^{27}\text{LiF-BeF}_2$) that was used in the MSRE. This salt is well understood and has a negative coolant void coefficient. Several other salts, such as the sodium-zirconium fluoride salts, have operational and cost advantages. Conceptual core design studies with more heterogeneous core designs are under way to determine if negative coolant void coefficients can be obtained for a wide variety of fluoride salts.

Economics

As shown in Table 2, preliminary overnight capital costs of the AHTR for several exit temperatures were determined relative to other higher-temperature reactor concepts [i.e., the S-PRISM and the gas turbine–modular helium reactor (GT-MHR)] based on the relative size of systems and quantities of materials. *This approach provides relative, but not absolute, costs.* Only the construction of multiple reactors provides reliable absolute costs. The lower capital costs are a consequence of several factors: economics of scale [2400-MW(t) reactor vs a 600-MW(t) or 1000-MW(t) reactor, passive safety in a large reactor system, and higher thermal efficiency.

Table 2 Comparison of estimated overnight capital cost (2002\$) of the AHTR-IT and AHTR-HT, as a percentage of the costs of the S-PRISM and GT-MHR [with multi-module output of 1145 MW(e)]^a

	<i>S-PRISM \$1681/kW(e)</i>	<i>GT-MHR \$1528 /kW(e)</i>
AHTR-IT \$930/kW(e)	55%	61%
AHTR-HT \$816/kW(e)	49%	53%

^a *The General Electric S-PRISM consists of four reactor modules, each producing 1000 MW(t) and 380 MW(e). The peak sodium temperature is 510°C. The General Atomics GT-MHR consists of four reactor modules, each producing 600 MW(t) and 285 MW(e). The peak helium temperature is 850°C.*

Research and Development (R&D)

About 80% of the R&D required for the AHTR is shared with that for helium-cooled high-temperature reactors. This includes fuel development, materials development, and Brayton power cycles. Many other areas are in common with facility design for liquid-metal reactors. The required R&D is sensitive to the peak coolant temperatures. Near 700°C, existing materials and fuels may be used. At 1000°C, major material development programs are required. Other areas that require significant R&D include (1) the reactor vessel insulation system, (2) optimization of core design, and (3) refueling and maintenance operations in the reactor vessel at 400 to 500°C. As a new concept, the AHTR is early in its development.

Table 1 Design characteristics of the 1970s MSBR

Net electric generation	1000 MW		Maximum core flow velocity	2.6 m/s
Thermal efficiency	44.4% (steam cycle)		Total fuel salt	48.7 m ³
Core height	3.96 m		²³³ U	1,500 kg
Vessel design pressure	5.2x10 ⁵ N.m ² (75 psi)		Thorium	68,100 kg
Average power density	22.2 kW/L		Salt components	⁷ LiF-BeF ₂ -ThF ₄ -UF ₄
Graphite mass	304,000 kg		Salt composition	71.7-16-12-0.3 mol%

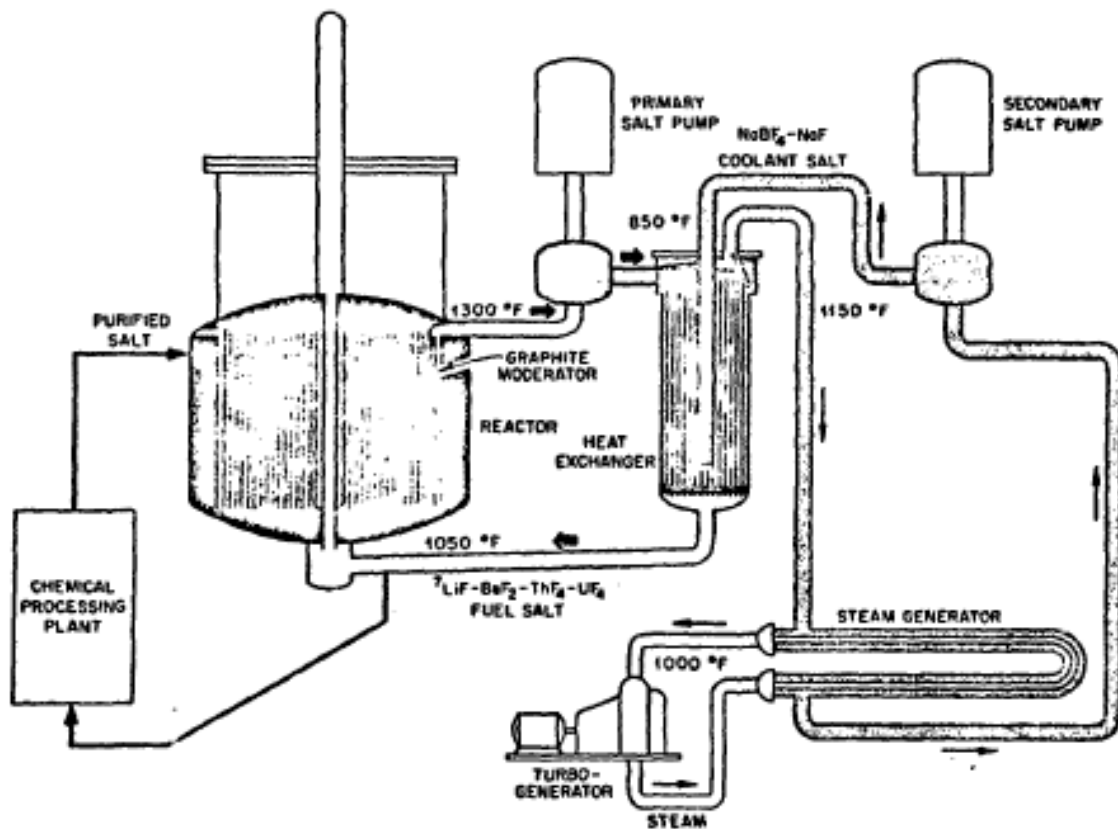


Figure 1 Single-fluid, two-region molten salt breeder reactor

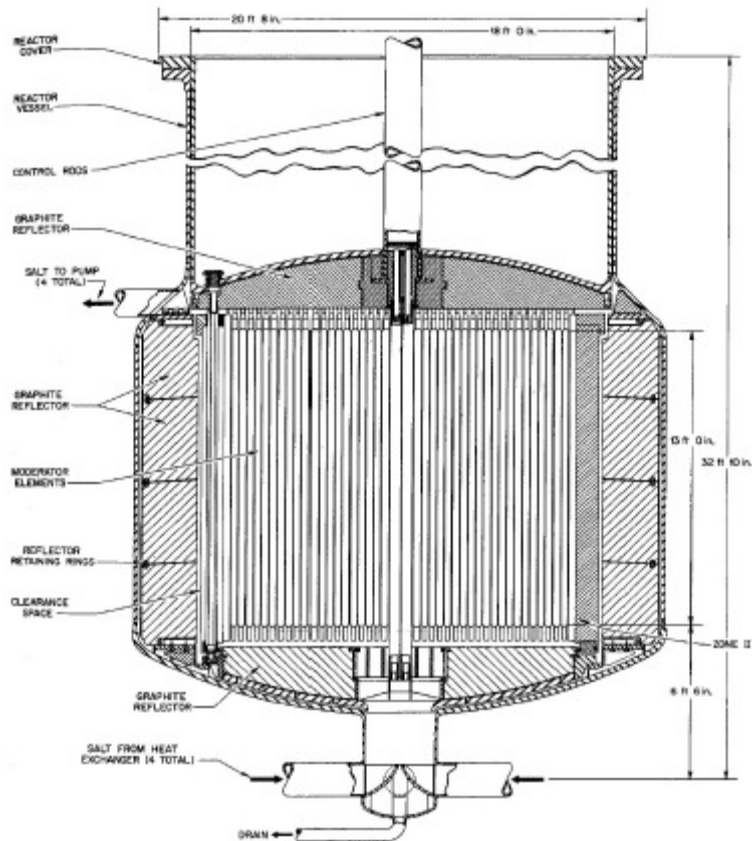


Figure 2 Cross section of the 1970s 2250-MW(t) MSBR vessel

Table 2 AHTR preconceptual design parameters

Power level	2400 MW(t)	Power cycle	3-stage multi-reheat Brayton
Core inlet/outlet temperatures (options)	900°C/1000°C 700°C/800°C 670°C/705°C	Electricity (output at different peak coolant temperatures)	1357 MW(e) at 1000°C 1235 MW(e) at 800°C 1151 MW(e) at 705°C
Coolant	2^7LiF-Bef_2 (NaF-ZrF ₄)	Power cycle working fluid	Nitrogen (Helium: long term option)
Fuel		Vessel	
Kernel	U-carbide/oxide	Diameter	9.2 m
Enrichment	10.36 wt% ²³⁵ U	Height	19.5 m
Form	Prismatic	Reactor core	
Block diameter	0.36 m (across flats)	Shape	Annular
Block height	0.79 m	Diameter	7.8 m
Columns	324	Height	7.9 m
Decay heat system	Air cooled	Fuel annulus	2.3 m
Volumetric flow rate	5.54 m ³ /s	Power density	8.3 W/cm ³
Coolant velocity	2.32 m/s	Reflector (outer)	138 fuel columns
		Reflector (inner)	55 fuel columns

Table 3: Results of corrosion tests in convection loops obtained by the ORNL

Alloy	T(°C)	Convection	Corrosion Rate (µm/year)
LiF-BeF ₂ -UF ₄			
Hastelloy N modified	676	Natural	0.5
	700		0.9
Hastelloy N Standard	660		1
LiF-BeF ₂ -ThF ₄ -UF ₄			
Hastelloy N modified	700		0.4
	704-566	Forced 3 to 6 m/s	3
	dT=55		1.5
Hastelloy N Standard	700	Natural	0.5

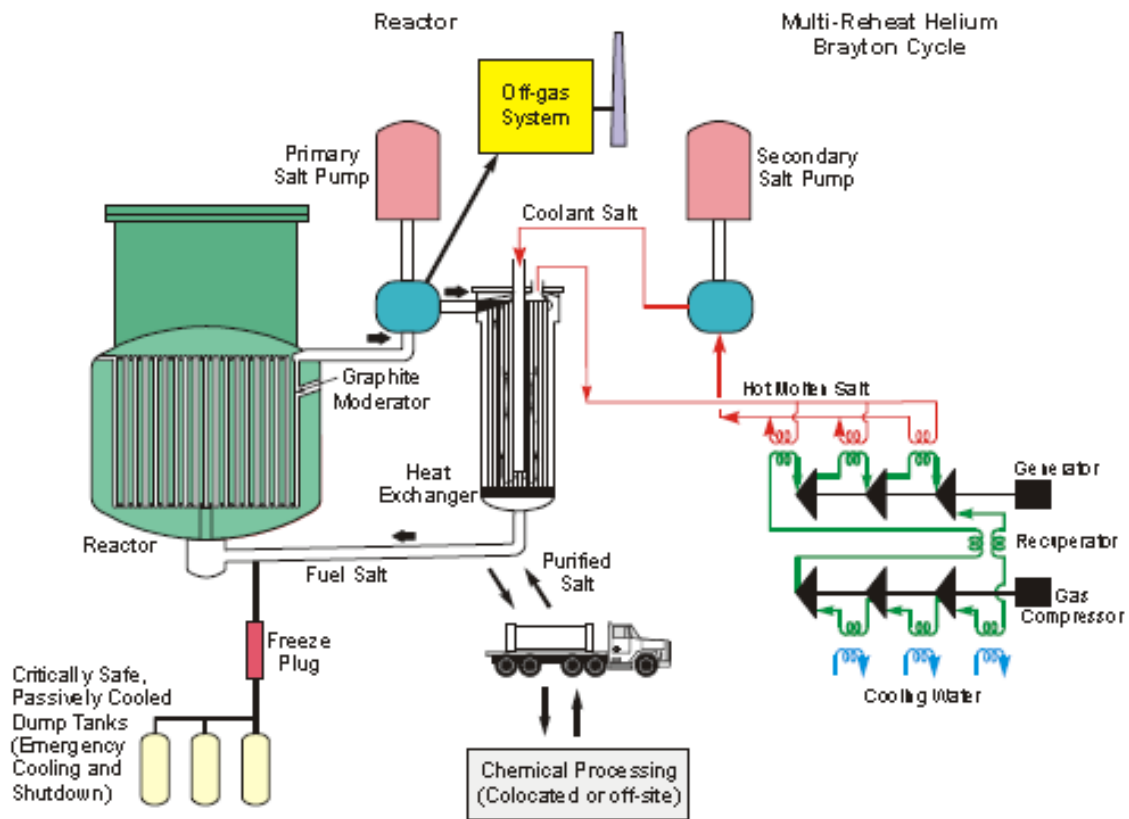


Figure 3 MSR with multi-reheat helium Brayton cycle

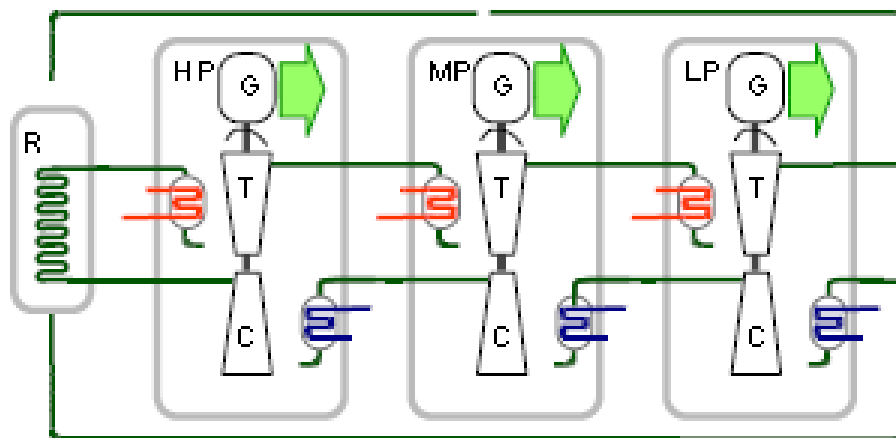


Figure 4 Schematic flow diagram for the reference three-expansion-stage Brayton cycle, using three PCU modules (HP, MP, and LP) each containing a generator (G), turbine (T), compressor (C), and heater and cooler heat exchangers, with a recuperator (R) located in a fourth vessel.

Table 4 Thermodynamic stability of components of molten salts and structural materials

Constituent	Free energy of formation at 1000°K (kcal/mol-F)	Cation thermal capture cross section (barns)
Majority Constituents (>99.9 mol %)		
^7LiF	-125	0.033 (^7Li)
MgF_2	-113	0.063
NaF	-112	0.52
RbF	-112	0.70
KF	-109	2.1
BeF_2	-104	0.01
ZrF_4	-94	0.18
^{11}BF	~ -95	0.05 (^{11}B)
AlF_3	-90	0.23
SiF_4	$\sim \text{XX}$	0.16
F	N	0.01
Structural Metal Constituents (Trace)		
CrF_2	-75.2	3.0
FeF_2	-66.5	2.5
NiF_2	-55.3	4.5
MoF_6	-50.9	2.5

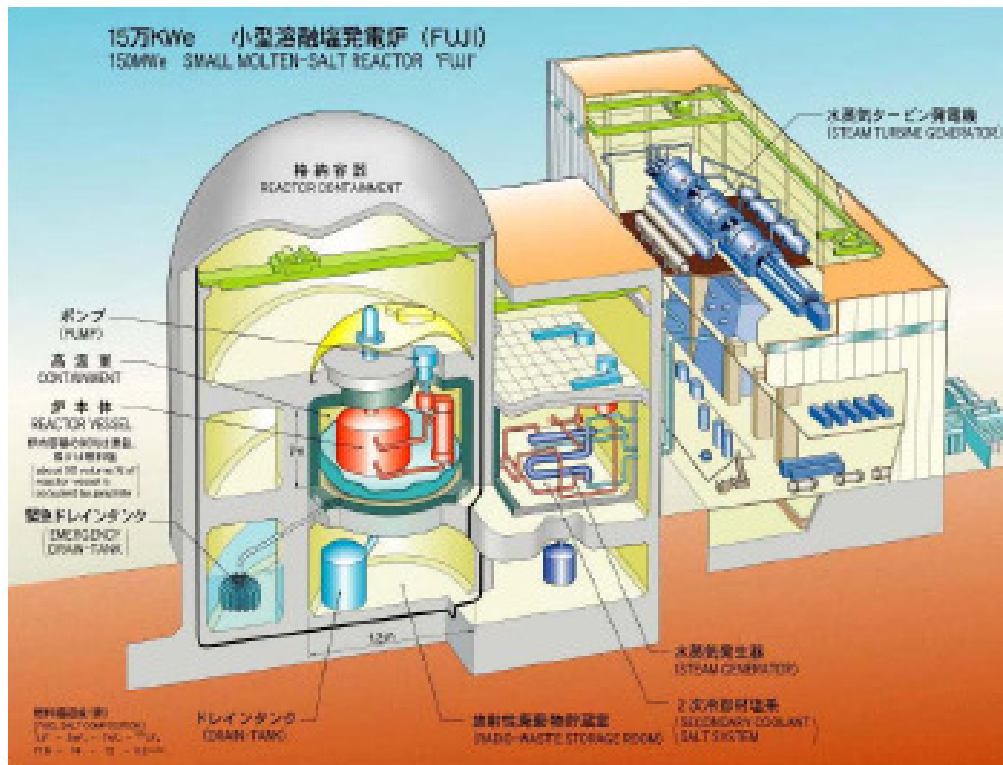


Figure 5 Full View of FUJI Molten-Salt Reactor

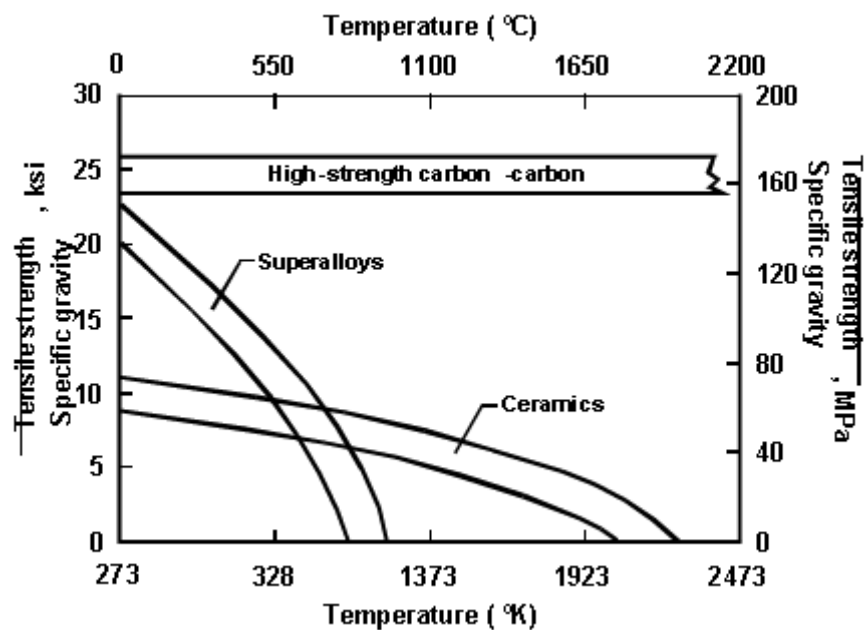


Figure 6 Strength of High-Temperature Materials Versus Temperature

Table 5 Physical properties of selected molten salts

Salt (mol%)	Form. Wt (g/mol)	Melt. Pt. (°C)	Density (g/cc), T(°C)	700°C Heat capacity (cal/g.°C)	Viscosity (cP), T(°C)	Thermal cond. (Watt/cm-°C)
Alkali-Fluorides (IA) : Nontoxic						
LiF-NaF-KF (11.5-46.5-42)	41.2	454	2.53-7.3E-4*T	0.45	0.04exp(4170/T)	0.006-0.01
LiF-RbF (43-57)	70.7	475	3.30-6.9E-4*T	0.284 (est)	0.021exp(4678/T)	~0.06
Alkali + Alkaline Earth Fluorides (IA + IIA)						
LiF-BeF ₂ (66-34)	331	458	2.28-4.884E-4*T	0.57	0.116exp(3755/T)	0.011
NaF-BeF ₂ (57-43)	441	360	2.27-3.7E-4*T	0.52	0.034exp(5164/T)	~0.01
Alkali + ZrF₄: Nontoxic and Low Tritium Yield						
NaF-ZrF ₄ (50-50)	104.6	510	3.79-9.3E-4*T	0.28	0.071exp(4168/T)	~0.01 (est)
NaF-KF-ZrF ₄ (10-48-42)	102.3	385	3.45-8.9E-4*T	0.26 (est)	0.061exp(3171/T) (est)	~0.01
Li-NaF-ZrF ₄ (42-29-29)	71.56	460	3.37-8.3E-4*T	0.35	0.0585exp(4647/T)	~0.01
Fluoroborates: Secondary Salt Candidates						
NaF-NaBF ₄ (8-92)	104.4	385	2.252-7.11E-4*T	0.36	0.877exp(2240/T)	~0.005
KF- KBF ₄ (25-75)	120.48	460	2.258-8.02E-4*T (est)	>0.32	Similar to KBF ₄	~0.005
RbF - RbBF ₄ (31-69)	151.25	442	2.494-8.7E-4*T	?	?	?
NaBF ₄	109.8	408	2.263-7.51E-4*T	0.36	0.0832exp(2360/T)	~0.005
KBF ₄	125.9	570	2.228-8.15E-4*T	0.32	0.0787exp(2406/T)	~0.005
RbF ₄	172.27	582	2.795-10.4E-4*T	?	0.0946exp(2280/T)	~0.005

^a Other mixtures of interest: NaF-RbF-ZrF₄ (8-50-42, mp = 400°C) and LiF-NaF-RbF (45-10-45, mp = 430°C), References: ORNL/TM-2316, ORNL/TM-4308, ORNL-4229; ORNL-4344, ORNL-4449, ORNL-4586, ORNL-4622, ORNL-4676; *Fluid Fueled Reactors* (1958); *Progress in Nuclear Energy Series 4*, Vol. 2, p. 140 (1960), *Nucl. Appl. Technol.* 8 (1970); *Nucl. Sci. Eng.* Vol. 71 p.200 (1963); R. DeWitt in *Phys and Chem.Liq.* Vol. 4 (no.2—3), pp.113-123 (1974).

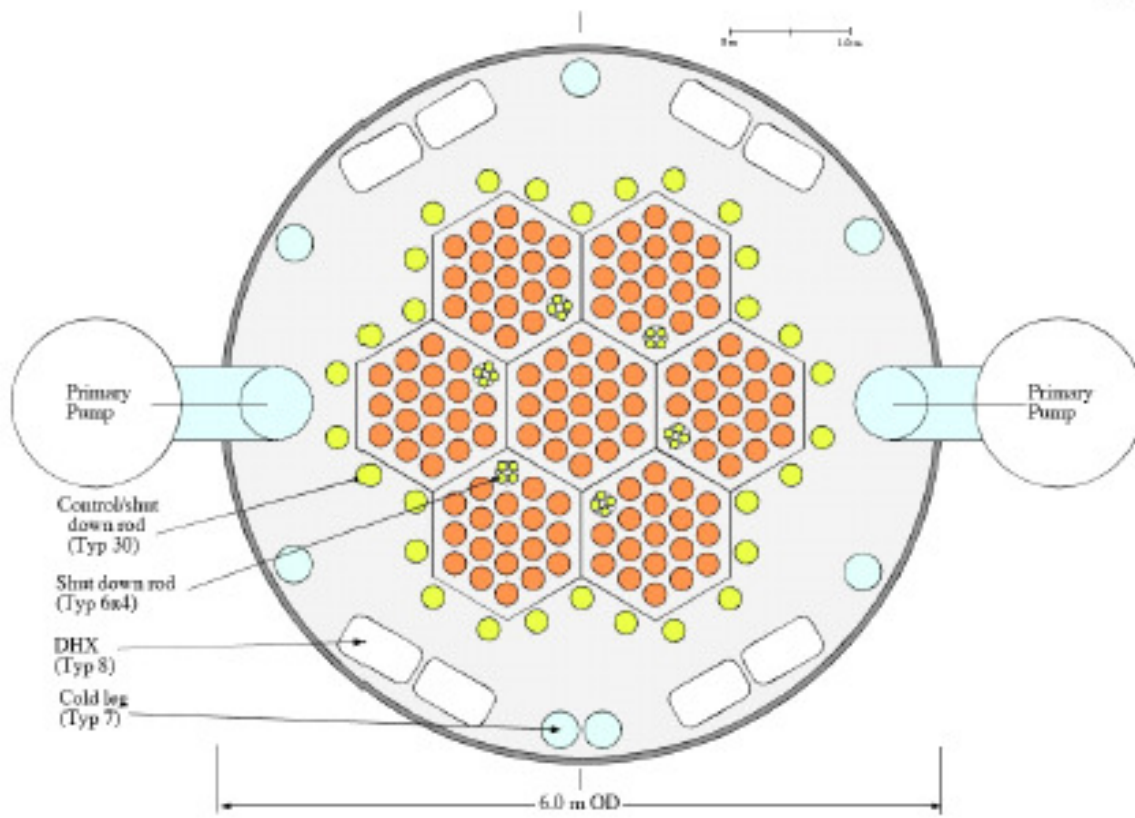


Figure 7 Reactor core cross section of 900-MW(t) PB-AHTR.

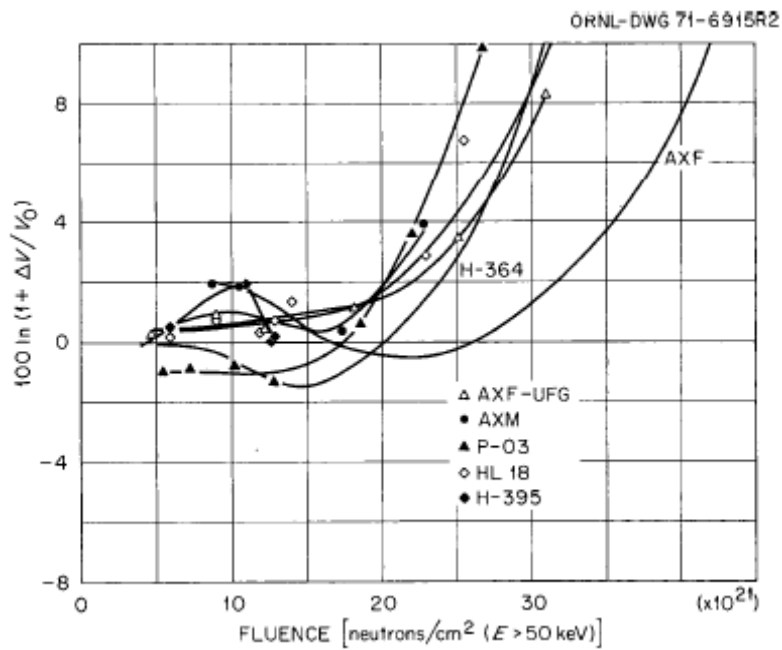


Figure 8 Volume changes for monolithic graphite Irradiated at 715°C(ORNL)

Table 6 Chemical compositions of Hastelloy N

Element	Content(% by weight) *	
	Standard alloy	Favored modified Hastelloy N (Ti modified) – (Nb modified)
Nickel	base	base
Molybdenum	15 - 18	11 - 13
Chromium	6 - 8	6 - 8
Iron	5	0.1 * *
Manganese	1	0.15 - 0.25 * *
Silicon	1	0.1
Phosphorous	0.015	0.01
Sulfur	0.020	0.01
Boron	0.01	0.001
Titanium and Hafnium		2 --> 0 (1976)
Niobium		(0 to 2) --> (1 to 2) (1976)
Cobalt		low enough

Single values are maximum amounts allowed. The actual concentrations of these elements in an alloy can be much lower.

These values are not felt to be very important. Alloys are now being purchased with the small concentration specified, but the specification may be changed in the future to allow a higher concentration.

Table 7 Chemical compositions of presented alloys, steels and standard Hastelloy N under investigation

Alloy or steel mark	Nb	W	Al	Si	P	Cu	Other elements
HN80MT	2.6	-	-	-	-	-	-
HN80MTY	-	-	1.03	0.01	0.006	-	-
12H18N10T	-	-	-	0.8	0.035	-	-
AP - 16	-	4.0 - 5.0		0.6	0.035	-	0.025 Ce
Hastelloy N	-	0.06	0.26	0.5	0.007	0.02	0.07 Cb

Alloy or steel mark	Ni	Fe	Cr	C	Mn	Ti	Mo
HN80MT	base	-	6.9	0.02	-	1.6	12.2
HN80MTY	base	0.06	5.5	0.03	0.01	0.83	12.3
12H18N10T	11 - 13	base	17 - 19	0.12	2.0	0.6 - 0.8	-
AP - 16	22 - 25	base	14 - 16	0.08	0.5 - 1	1.4 - 1.8	-
Hastelloy N	base	3.97	7.52	0.05	0.52	0.26	16.28

Table 8 Summary of corrosion testing results for salts without uranium

Loop	Alloy	Salt	Time of exposure [h]	Temp [°C]	Corrosion depth [μm]
116	316SS	FLiNaK + NaK	500	815	4
119	316SS	FLiNaK + NaK	500	815	2
347	Inconel	50NaF-50ZrF4	3000	815	11
518	Inconel	NaF-ZrF4	3000	815	11
346	Inconel	50NaF-50ZrF4	2000	815	9
519	Inconel	NaF-ZrF4	2000	815	12.5
78	Inconel	FLiNaK	1000	815	13
	Inconel	NaF-ZrF4	1000	815	3
278	Inconel	NaF-ZrF4	1000	815	5
399	Inconel	NaF-ZrF4	1000	815	10
	Inconel	60NaF-40ZrF4	1000	815	5
	Inconel	50NaF-50BeF2	1000	815	8
	Inconel	70NaF-30BeF2	1000	815	6
	Inconel	24LiF-53NaF-23BeF2	1000	815	5
	Inconel	36LiF-49NaF-15BeF2	1000	815	3
	Inconel	74LiF-26ThF4	1000	815	6
517	Inconel	NaF-ZrF4	822	815	5.5
337	Inconel	NaF-ZrF4	575	815	8
214	Inconel	FLiNaK + NaK	500	815	3
230	Inconel	36NaF-18KF-46ZrF4	500	815	10
348	Inconel	50NaF-50ZrF4	500	815	5.5
	Inconel	FLiNaK	500	815	7
934	Inconel	60NaF-40ZrF4	500	815	5
935	Inconel	60NaF-40ZrF4	500	815	5
	Inconel	NaF-BeF2	500	815	10
	Inconel	LiF-NaF-BeF2	500	815	5
246	Inconel	52NaF-48ZrF4	500	815	8
262	Inconel	57NaF-43BeF2	500	815	9
277	Inconel	50NaF-50ZrF4	500	815	5
276	Inconel	NaF-ZrF4	500	815	8
277	Inconel	NaF-ZrF4	500	815	4
336	Inconel	NaF-ZrF4	500	815	6
341	Inconel	NaF-ZrF4	500	815	5.5
342	Inconel	NaF-ZrF4	500	815	6
516	Inconel	NaF-ZrF4	500	815	6
338	Inconel	NaF-ZrF4	500	815	6
411	Inconel	NaF-ZrF4	250	815	4.5
410	Inconel	NaF-ZrF4	100	815	4
400	Inconel	NaF-ZrF4	50	815	3
1181	Inconel	71LiF-29ThF4	8760	732	6.5
1239	Inconel	71LiF-16BeF2-13ThF4	8760	732	7.5
9377-6	Inconel	71LiF-16BeF2-13ThF4	13155	704	13
1188	Inconel	35LiF-27NaF-38BeF2	8760	677	9
1210	Inconel	71LiF-29ThF4	8760	677	5
1235	Inconel	71LiF-16BeF2-13ThF4	7789	677	4
1214	Inconel	FLiNaK	4673	677	13
1169	Inconel	71LiF-29ThF4	1000	677	1
1177	Inconel	71LiF-29ThF4	1000	677	1.5
1173	Inconel	58NaF-35BeF2-7ThF4	1000	677	4
1176	Inconel	58LiF-35BeF2-7ThF4	1000	677	1
1234	Inconel	71LiF-16BeF2-13ThF4	1000	677	1
9344-2	Inconel	FLiNaK	8760	649	8
9344-2	Inconel	FLiNaK	8735	649	8
1172	Inconel	35LiF-27NaF-38BeF2	1000	607	2
1175	Inconel	FLiNaK	1000	607	1

Loop	Alloy	Salt	Time of exposure [h]	Temp [°C]	Corrosion depth [μm]
MSRE	INOR-8	67LiF-33BeF ₂	26000	649	0
9354-3	INOR-8	35LiF-27NaF-38BeF ₂	19942	649	0
1194	INOR-8	FLiNaK	1000	607	0
1195	INOR-8	35LiF-27NaF-28BeF ₂	1000	607	0
LDRD	INOR-8	FLiNaK	3048	815	0.1
1209	INOR-8	71LiF-29ThF ₄	8760	732	0
1216	INOR-8	58LiF-35BeF ₂ -7ThF ₄	8760	732	1
1240	INOR-8	71LiF-16BeF ₂ -13ThF ₄	8760	732	0
MSRP7	INOR-8	71LiF-16BeF ₂ -13ThF ₄	20000	704	1
MSRP8	INOR-8	58LiF-35BeF ₂ -7ThF ₄	9633	704	0
15A	INOR-8	73LiF-2BeF ₂ -25ThF ₄	39476	677	0.05
1208	INOR-8	FLiNaK	8760	677	1
1190	INOR-8	58NaF-35BeF ₂ -7ThF ₄	8760	677	1
1233	INOR-8	71LiF-16BeF ₂ -13ThF ₄	8760	677	0
1213	INOR-8	71LiF-29ThF ₄	3114	677	0
15	INOR-8	73LiF-2BeF ₂ -25ThF ₄	2003	677	0
1165	INOR-8	FLiNaK	1340	677	0
1164	INOR-8	58NaF-35BeF ₂ -7ThF ₄	1000	677	0
1221	INOR-8	71LiF-29ThF ₄	1000	677	0
1228	INOR-8	71LiF-16BeF ₂ -13ThF ₄	1000	677	0
MSRE	INOR-8	67LiF-33BeF ₂	26000	649	0
9354-3	INOR-8	35LiF-27NaF-38BeF ₂	19942	649	0
1194	INOR-8	FLiNaK	1000	607	0
1195	INOR-8	35LiF-27NaF-28BeF ₂	1000	607	0

CONCLUSIONS

Based on the preceding review of nuclear, physical, chemical, and economic properties of candidate salts for the AHTR, the following conclusions were reached:

Salts composed of low-atomic-weight constituents ("light" salts) possess superior heat transfer metrics for use as the AHTR coolant. Heavier salts are also relatively good coolants and would likely prove acceptable for design purposes.

Analysis indicates that the key reactivity coefficients (and their net effect) that control response to transients are more strongly affected by parameters associated with the fuel-block design (coolant volume fraction, poison level, and distribution) than by the identity of the particular salts under consideration. A computational framework was developed to evaluate these factors and was used to evaluate the various candidate salts. It appears that acceptable fuel element designs can be found for most of the salts used in this study.

Activation levels in AHTR candidate coolants appear to be acceptable from both an operational and long-term disposal standpoint. Only the LiF-BeF₂

salts are very low-activation materials that support minimal shielding requirements. The other salt coolants have operational characteristics similar to those of sodium-cooled reactors. The disposal of all of the salt candidates as LLW after 60 years of operation and 10 years of cooling should be possible, although some simple pre-treatment may be required for certain coolants.

No consensus exists to select a particular salt based on its corrosion behavior with high temperature alloys.

A number of the ZrF₄-containing salts appear to offer the best potential for achieving a low cost coolant. The economic basis for these judgments is very important for the selection of a candidate and needs to be refined.

RECOMMENDATIONS

The selection of a suitable coolant salt should be based on numerous factors, including chemical, physical, nuclear, metallurgical, and economic factors. It is evident from past decades of experience that fluoride melts have an established advantage over the few other coolants that had been considered previously for extreme-temperature service (>700°C). The following remarks are directed primarily toward selection based on chemical factors that relate to corrosion, with the understanding that the overall assessment will need to account for other factors.

Proper selection of a coolant salt based on chemical differences is based largely on the acid-base properties of the combination, as described in Sect. 6. Both predictions and measurement of the container-metal-fluoride equilibrium concentrations are higher in basic salts as compared with neutral or acidic media. Some corrosion loop experience tends to corroborate this observation.

Unfortunately, however, no systematic study of such a phenomenon has been made during these experiments.

A neutral or slightly acidic salt melt would be predicted to be the most advantageous with respect to corrosion behavior. However, basic salt melts tend to have significantly lower vapor pressures and lower viscosities, and these properties might present a problem (for example, ZrF₄ and BeF₂). Therefore, any selection of a coolant based on chemical considerations must, necessarily, be a compromise of all factors that might affect performance.

In this regard, it is recommended that two types of salts should be studied in the future:

- (1) Salts that have been shown in the past to support the least corrosion and are neither strong Lewis acids nor strong Lewis bases (i.e., "neutral"). Salts containing BeF₂ and ZrF₄ in the concentration range 25–40 mol % fall into this category.

(2) Salts that provide the opportunity for controlling corrosion by establishing a very reducing salt environment. The alkali-fluoride (FLiNaK, FLiNaRb) salts and the BeF₂-containing salts fall into this category.

It is too early to make final recommendations for exact salt compositions, since the particular salt composition to be chosen will need to be determined from a carefully conducted trade study that balances the various selection factors for a particular reactor design.

CANDIDATE SALTS

Separate recommendations are made for work in the future with respect to (a) analysis and computations, and (b) experimental work. These recommendations do not constitute a final selection, rather they identify important candidate salts that exhibit important properties that should establish useful boundaries for future evaluations.

Further computational analysis of AHTR candidate salts is recommended on the following basis:

1. ZrF₄-salts that have the potential for low cost and special properties:
 - NaF-ZrF₄ and related ternary mixtures :
 - i. NaF-ZrF₄ (59.5-40.5) *potentially least expensive*
 - ii. 7LiF-NaF-ZrF₄ (42-29-29) *very low vapor pressure*
 - iii. RbF-NaF-ZrF₄ (34.5-24-41.5) *low melting point (420°C)*
2. BeF₂-salts that have superior nuclear properties and heat-transfer performance:
 - NaF-BeF₂ *low melting point salt (340°C)*
 - 7LiF-BeF₂ *neutron transparent salt, very low activation*

All alkali fluoride compositions require significant amounts of 7Li, and the mixtures with the most favorable heat-transfer properties (FLiNaK) have the poorest neutronic performance. The BeF₂ and ZrF₄ salt systems both have compositions that melt below 400°C and do not contain lithium, whereas the alkali fluorides do not.

Further experimental work is recommended with the following salts:

1. NaF-ZrF₄ and related ternary mixtures :
 - i. NaF-ZrF₄ (59.5-40.5) *potentially least expensive*
 - ii. 7LiF-NaF-ZrF₄ (42-29-29) *very low vapor pressure*
 - iii. RbF-NaF-ZrF₄ (34.5-24-41.5) *lowest melting point*
2. Alkali fluoride salts (e.g., FLiNaK) as a reference salt. These salts will establish the bounding corrosion response, and permit corrosion studies under a wide range of redox conditions.

The corrosion and properties database for BeF₂ salts is rather extensive. BeF₂ salts are expensive, and they impose significant costs in conducting tests. It is likely that much of the information needed for work at temperatures above 750°C can be gathered initially with other inexpensive

and less hazardous salts. This leaves the option open for a future selection of BeF₂ salts, should that option prove to be the best one.

FUTURE WORK

The database for salt thermal conductivity and heat capacity needs significant improvement.

In particular, the properties database for ZrF₄-salts is in question and merits further investigation.

An integrated materials design and testing strategy will be required to make the optimal choice of salt based on corrosion metrics. It is recommended that the salt-chemistry, corrosion chemistry, and alloy-selection studies be conducted as a joint effort.

Although there remain a number of technical issues with respect to molten salt coolants, a better understanding of economic factors for the production of industrial-scale quantities of these materials is even more urgent. It is recommended that a study be done to place the cost of salts for an industrial deployment on a sound footing. At present, only NaF can truly be considered a commodity chemical for the production of salt.